Engineered Passive Barrier Systems for Mitigating Landfill Gas Emissions

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ABSTRACT

The objective of this study was to provide an alternative methanotrophic system aimed at filtering out methane and other gases emitting from landfills and to reduce their release into the atmosphere in arid land, in a passive way. Methane and other landfill gases are oxidised and converted into carbon dioxide and other less harmful by-products through microorganisms present in soils (methanotrophic bacteria) and in the presence of oxygen. The inherent low air diffusion in the top cover layer of landfills, compounded by the low permeability soils existing in desert countries, such as Kuwait, renders the use of conventional passive mitigation systems unusable in these countries. Based upon these facts, a comprehensive literature review revealed that the abundance of the research was dedicated to finding better mitigation systems to deliver oxygen to the landfills' covers through the top surface. Even by designing a structure or experimenting with cover materials, the air delivery of these passive filter systems is still dependent upon the natural atmospheric and molar diffusion into the soil, which, even insufficient and inadequate delivery mechanism in normal circumstances, could not be used in landfills located in arid land environment. Through a series of batch experiments, oxygen was found to be the most important and limiting factor that surpassed the other factors needed by the methanotrophic bacteria for any landfill and particularly for landfills located in arid lands. The batch test also highlighted the importance of supplying oxygen in an adequate and continuous flow, and revealed that a time lag of three to four days could occur before the methanotrophic bacteria could regenerate and commence converting methane gas. In addition, a continuous flow reactor experiments (CFR) confirmed that poor nutrient desert sand and soil cover could not support efficient methane elimination, making it necessary to supplement these covers with nutrient-rich amendments. The CFR experiments also showed that a supply of oxygen in levels deeper within the soil layers has a much higher methane oxidation rates than the conventional surface oxygen supply, reaching more than 65% average rate of oxidation. Based on these findings, and on the particular circumstances in arid lands, particularly the dust-laden environment of Kuwait, a passive concept system was proposed and a modelled numerically to compare with results of field a trial data, which showed advantage of this passive system over conventional systems. A mitigation scheme was also recommended to control environmentally harmful gases from release into the atmosphere from landfills located in the arid land of Kuwait.

Declaration

The content of this thesis is solely my responsibility, and the original work herein is my own, except where specified otherwise in the text. Neither the thesis nor any of the original work comprising it has been submitted to this or any other institution for consideration for a higher degree.

Signed…….

Date.....

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1.1 Introduction

Human activities and their impact on the environment have become a great concern to the world community at this present time. These activities are driven by population explosion through the years, showing exponential and uncontrolled rates of human growth, which in turn, has influenced the earth's environment in an unsustainable way. Figure 1.1 shows population expansion during the past millennium since 10,000 BC to the present time (Marchetti, 1996; UN, 1992), in which humans have multiplied steadily at an alarming rate, specifically in the past 2000 years since the creation of the earth itself. The population went from several millions before the time of Christ reaching 7 billion now, and is expected to reach 9 billion in 2050 (UN, 2010).

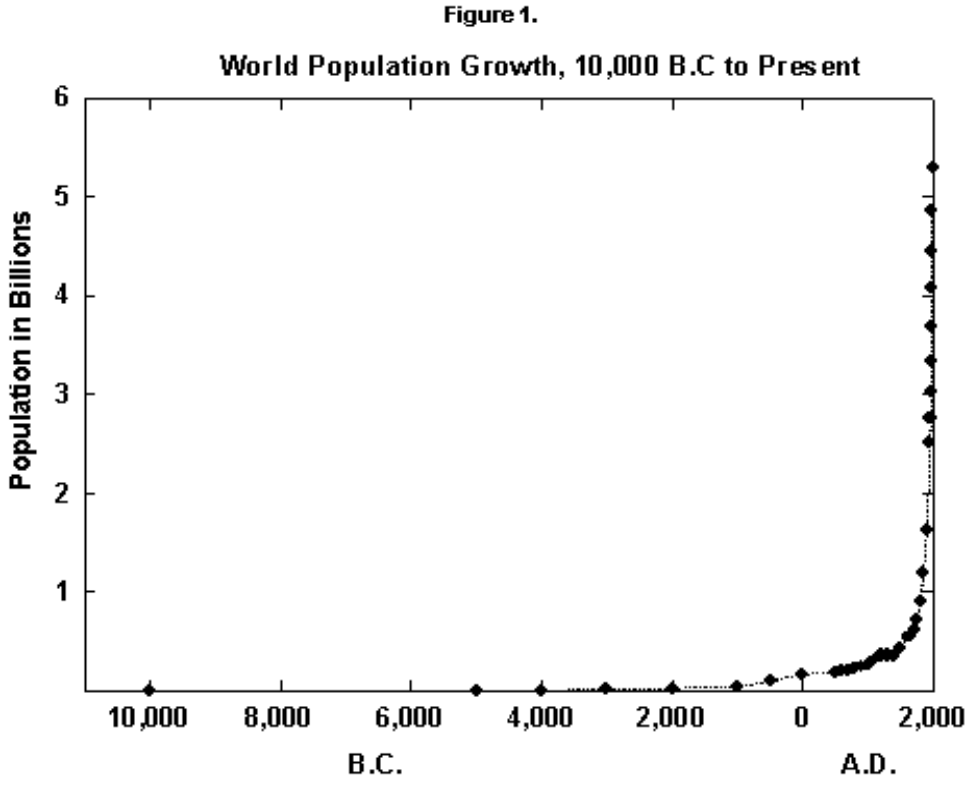

Figure 1.1: Human rate of growth during 12000 years of time (sources: Marchetti, 1996; UN, 1992)

These human activities, which include agriculture, coal mining, biomass burning, gas and petroleum production, wastewater treatments, industrial processing and production, and quite simply, the activities of 7 billion humans on the planet, have had such a profound impact on the earth's environment. Municipal solid waste production is another major human activity that, when degraded, produces high amounts of methane gas pollutants competing highly with the other high producing activities. These human solid waste products, for a lack of a better means of disposal, are buried in the crust of the earth's surface in enclosures, referred to as landfills. (Hrad, et al., 2012, Abushammala, et al., 2014, Reddy, et al., 2014, Huber-Humer and Lechner, 2014).

In landfills, discarded biological and waste materials are collected daily. They piled up, and when fermented enough, they produce substantial amounts of methane gas. The emitted methane production from these landfills was estimated to exceed 10–20% of the total methane produced per year globally in some estimates (He, et al., 2008, Kettunen, et al., 2006, Jugnia, et al., 2008). It also could reach up to 37% of the total methane produced per year from landfills in the USA, according to Stern et al., (2007) and Xu et al., (2014). In contrast, it is likely that some of the northern hemisphere areas could experience warming up to 40% more than the mean global warming average in cold seasons due to greenhouse gas emissions (Houghton et al, 2001). These estimates are projected using different models to simulate and predict methane production in landfills, and on using assumed input parameters, produce different predictions for different sets of assumed initial parameters (Section E.4 of Appendix E). In fact, methane production from landfills is one among the top largest emitters globally generated from human activities, along with agriculture and fossil fuel emissions (Forster et al., 2007). These percentages could translate to 20–70 Tg of methane (CH_4) emitted into the atmosphere yearly (Khalil et al., 1989), an indication of CH_4 escalation from only 700 ppb in 1750 to more than 1774 ppb in 2005 (IPCC, 2007). Other studies have estimated global methane production to reach more than 300–400 Tg CH_4 as a result of human activities (Blake et al., 1988). Moreover, methane concentration in the atmosphere has increased from 0.35 to 1.7 ppm in the past 18000 years; while it has increased from 0.75 to 1.7 ppm in just the last 300 years, and is

estimated to increase further to range between 2.1 and 4.0 ppm in 2050 (Blake et al., 1988; King, 1994).

Based on the 1996 and 2006 IPCC estimates and guidelines (Bogner et al., 2007), the global production of methane from landfills as a result of biodegradation, is estimated to reach 500–800 million metric tons of carbon dioxide equivalent per year ($MMTCO_2$-eq), and is projected to reach up to 2900 $MMTCO_2$-eq/year in 2050. A comparison of the level of global methane production, due to wastes decompositions in landfills with other highly producing industries, is shown in Table 1.1. Methane effects on the atmosphere are significantly higher than that of carbon dioxide, as it has 72-folds greater global warming potential (GWP) over a 20-year period and 25-folds greater GWP over a 100-year period in comparison to carbon dioxide (IPCC, 2007). This difference in GWP for methane in the short- and long-terms could be attributed to the rapid degradation of methane compared to that of carbon dioxide in the atmosphere. Another estimate suggested that methane has a slightly smaller effect than the aforementioned estimates, having only 23-folds greater GWP than carbon dioxide (Houghton et al., 2001), which nonetheless, is still significant. Its potent effect is attributed to the fact that mole per mole methane has the capacity to absorb infrared irradiation for wider range of wavelengths than carbon dioxide does (Lelieveld et al., 1993). These alarming indicators are evolving continuously, despite the effort to utilise, contain, or burn methane gas into the atmosphere, using processes that can reduce its potency from its present effect as CH_4, to lower level as carbon dioxide (CO_2).

Much of the global production of these greenhouse gases emanates directly from landfills, with a composition of 35–60% methane and 30–55% carbon dioxide (Rasi et al., 2007). Production of landfill gases (LFG) occurs naturally from the slow biodegradation of biological materials and other materials in landfills over many decades. In this process, an increase in pressure inside these landfills would generate an advective flux of these gases vertically within the landfill, because they exceed atmospheric pressure, hence, escaping into the atmosphere as a natural process (Kjeldsen, 1996). Factors such as the type of waste, biological amount, volume, temperature, moisture, oxygen presence, etc., can have a direct effect on

methanogenic microorganisms to ferment this biological material. Methane production occurs strictly in the absence of oxygen and in an anaerobic condition.

Industry	Methane Emission Processes	Global Methane Production level ($MMtCO_2$-eq)
Oil and Gas Systems	Emitted during normal operations, routine maintenance, and system disruptions in the oil and natural gas industry	**1354.42**
Landfills	Produced through the decomposition of organic waste under anaerobic conditions typically found in landfills and large dump sites	**760.63**
Wastewater	Produced by decay of organic material in wastewater as it decomposes in anaerobic environments.	**594.04**
Coal Mines	Emitted from active and abandoned underground mines and surface mines, and as a result of post mining activities including coal processing, storage, and transport	**407.56**
Agriculture (manure management)	Produced from decomposition of livestock and poultry manure stored or treated in systems that promote anaerobic conditions (e.g., liquid or slurry in lagoons, ponds, tanks, or pits).	**243.95**

Table 1.1: Global methane emission by most polluting industries (Source: Global Methane Initiative, 2015)

Methane production in landfills occurs almost immediately when materials of organic nature are deposited in a landfill. This is because the organic matter usually undergoes some decomposition during transport, depending on many factors such as storage time, humidity, and temperature inside the stockpiles, as well as the transporting vehicles. Unfortunately, this part in the process of methane production has not, as yet, been researched extensively. More importantly however, is the time that is spent filling landfills with wastes prior to actively extracting gas by any controlling methods. This is because, traditionally, landfills are kept in a waiting mode each day during filling,

until they are filled with waste, and then covered, and only after closure, that controlling methods are put in place for long-term control of methane emissions. Extracting methane for heating, electricity, or for flaring are some of the traditional processes installed on landfills, and when these systems become unattainable due to the decline of gas release, landfills are then closed and abandoned, despite the continuity of gas emission into the atmosphere. Significant amounts of gas are left in landfills to escape or migrate into the soil, if not subjected to any form of control methods. Huber-Humer et al. (2008) found out from actual landfill experiments, that a significant proportion of gas emission is not captured by any of the meaningful control treatments, and that the amount of uncaptured gas could reach up to 40–50% during the lifetime of the landfills, as demonstrated in Figure 1.2.

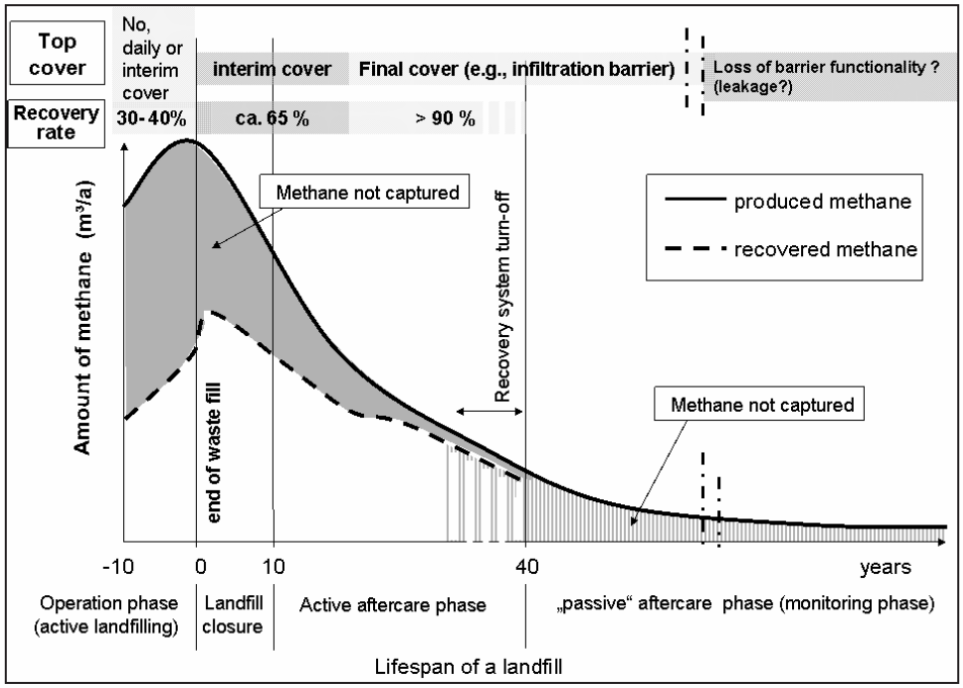

Figure 1.2: Gas extractions during the life span of a landfill (source: Huber-Humer et al., 2008).

From this graph, the amount of methane left to escape is quite significant, particularly during the active landfilling stage, where wastes are still fresh. The other significant part left to escape is when the landfills' controlled collection methods are turned off, in which stage, the aged landfill becomes leaky and damaged, but which still produces harmful environmental gases, due to time and environmental elements.

During the life span of landfills, microorganic bacteria exist abundantly and take different forms, shapes, and actions. These microorganisms are important in decomposing all sorts of matters, from hard minerals to soft organic materials. Recent international regulations, such as carbon production taxation, as well as economic and environmental impact, have encouraged and stimulated the attention and interest in the action of methanotrophs in the soil and other microorganisms that can be used to convert biological and other materials of the waste in a natural and passive way.

1.2 Landfill gas emission in arid climate

Many high-income developing countries produce high volumes of waste that predominantly end up in landfills, with virtually no means of controlling the resulting emissions. Table 1.2 shows the quantity of solid wastes generated per day, per capita in some developing nations (Asfari, 2002), indicating a production of high volumes of waste among high-income developing nations in comparison to others. Of particular interest is the amount of waste generated in Kuwait, where 1.8 kg/d/capita is produced — the highest of any nations in that study. This amount of production, compounded by the small area of this country accounting for only 17820 km^2, makes it difficult to find new sites for landfills. With this small area available, surprisingly, the country has a high number of landfills scattered around the urban areas. The numbers exceed 18 landfills (some are now closed), to serve approximately 4 million inhabitants, receiving more than 500 tons of waste a day. The worst part is that the area of the country marked for urban usage is only 25%; while the rest is owned by petroleum, private companies, and occupied by government and military installations, thus obliging the municipality to place these landfills close to the residential areas (The Industrial Bank, Kuwait 2010). Furthermore, the landfills are managed poorly, with no recycling facilities, and have no gas control systems installed on them. Due to these poor landfill conditions, fires flare occasionally; subsequently, posing serious concerns not only relative to the management of landfills in Kuwait, but also more importantly, on the health of the residents nearby, because of scarce land availability.

Country	Municipal waste (kg/Capita/day)	Country	Municipal waste (kg/Capita/day)
Bahrain	1.6	Qatar	1.3
Egypt	1.2	Saudi Arabia	1.3
Jordan	0.9	Syria	0.5
Kuwait	1.8	Tunis	0.6
Oman	0.7	UAE	1.2
Morocco	0.33	Yemen	0.45

Table 1.2: Comparison of solid waste discarded in landfills in some developing nations (Source: Asfari, 2002)

A comparison of solid wastes discarded in landfills of some citiesF of developing nations against those discarded by Kuwait is shown in Table 1.3, indicating a production rate similar to that of the high percentage of foodstuff discarded by high-income countries. The table shows that an average of 50–60% of the produced solid wastes is organic (Asfari and Mashan'n, 2002). In particular, Kuwait has high organic waste production, which is dumped in landfills that were contained in a high temperature climate, producing free gases to the atmosphere.

Location Category (%)	Aden	Aleppo	Amman	Bahrain	Cairo	Kuwait	Riyadh	Tunis	Mumbai
Food Waste	57.1	59.4	54.5	59	67	50	34	68	58
Paper and boards	10.7	13.1	14	12.8	18	20.6	31	10	10
Plastics	10.8	11.5	13.2	7.44	3.4	12.6	2	11	11
Metals	5	0.8	2.4	2.05	2.2	2.6	16	4	2
Glass	2.7	7.6	2.8	3.39	2.5	3.3	3	N/A	3
Wood	N/A	0.5	N/A	N/A	N/A	4.8	10	N/A	N/A
Textile, Rubber and Leather	5.6	3.7	4.7	6.92	0.5	4.8	2	2	N/A
Yard Trimmings	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A
Others	8.1	3.4	8.4	8.4	6.4	1.3	2	5	16

Table 1.3: Solid waste percentages discarded in landfills in some cities of developing nations (Source: Asfari and Mashan'n, 2002)

1.3 Objectives

Waste deposited in landfills in Kuwait contains almost all types of wastes, including municipal solid wastes (MSW) and industrial wastes all dumped in the same dumping grounds, which are not even designed for sanitary fillings. The industrial wastes comprise liquids and sludges produced by factories, dairy and food factory products, oil and industrial hazardous products, factory metal discards, contaminated soils, hospital and private laboratories wastes, all types of batteries, and many uncategorised wastes, which in total constitute approximately 44% of the total wastes discards (Al-Yaqout et al., 2005). These assortments of wastes interact chemically and biologically, producing all kinds of gases. Unfortunately, studies characterising these gases are scarce. However, a study conducted by Al-Yaqout et al. (2005) on Al-Qurain landfill in Kuwait showed that methane gas was the major constituent of the gases emitted from that landfill. Methane production was 52.47%, and carbon dioxide was 36.14%, all producing at rates of 149–567 ml/min through a six-inch diameter boreholes immersed at 6–3 m depths. Although these rates were not high, Winthesier (1996) has reported that rates of methane production in arid lands could produce gases in quantities much more than the quantities produced by landfills in temperate or humid climates.

High outside temperatures, little rain precipitation, and poor nutritional contents of the soil cover material existing in arid landfills, suggesting that these environments would hinder oxidation, however, did not prevent biological degradation of the materials from happening in these landfills (Al-Yaqout et al. 2005). While these conditions pose a challenge, measurable amounts of gases are being produced from landfills existing in Kuwait. The ambient temperatures surrounding these landfills, for the Kuwaiti example, range from 36 to 48°C, lasting for about seven months of the year, and 18–25°C the rest of the months; while rain precipitation does not exceed more than 10 cm a year (Weatherspark, 2014). On the other hand, the inside temperature measured for Al-Qurain landfill had been found to range between 23 and 35°. The inside high moisture contents could have been due to the high volumes of liquids deposited in the

landfills, eventually converted into vaporous moisture gas, diffusing throughout the soil (Al-Yaqout et al., 2005). The challenge in these arid climate lands, albeit, is the type of cover soil that must be used on these landfills. In practice, earth soils dug from the site were used again as covers, without any form of amendments added. The soil characteristic in Kuwait is highly impermeable clay and sand (called Gutch locally), having a low hydraulic conductivity in the range of 4.0 $x10^{-7}$ cm/ s, and pH in the range of 7.8 to 8 (Al-Yaqout et al., 2005; Al-Yaqout and Townsend, 2004). The real challenge, with regard to the landfill cover in Kuwait, besides the low hydraulic conductivity and poor nutrient contents, is the amount of dust fallout covering the surface of the landfills due to frequent sandstorms. The rate of dust fallout was calculated to reach 278 T km^{-2}, falling on an average of 255.4 d of the year (70% of the year) (Al-Dousari et al., 2014). Krishna and Suresh (2016) in their recent study have found out that there is a decrease in the permeability of the soil with the increase in the addition of stone dust. Therefore, it is anticipated that the continuous dust fallout could clog the fine pores and voids of the desert surface, further aggravating the poor natural permeability of the soil. These unique arid environments have rendered ineffective any conventional or new landfill cover technologies.

With these challenging arid climate conditions, existing particularly in Kuwait, apparently, conditions are suitable for methanogenic bacteria to biodegrade organic matters, but not quite right for methanotrophic bacteria to assimilate methane. Due to insufficient air (and oxygen) diffusing through the landfill covers, poor vegetation, and low organic nitrates for bacterial activities, methane is left to escape unabated. The use of conventional methods to capture methane for use is subject to economic viability, which no private company seems to want to take (The Industrial Bank, Kuwait 2010). In addition, recent bio-cover technologies require biological amendments to be installed within or on the cover layers. The biological materials for amendments (i.e. compost materials) require sorting and composting facilities for the biological matters, which again are neither available nor economical to install, considering that these materials, once produced, may not be useful in the country. Consequently, due to international pressure in the form of carbon taxation, the authorities in Kuwait are now

trying to discover new ways and means to control its carbon emissions for their closed and old landfills.

The goal of this study, therefore, is to propose a mitigation system suitable for reduction of methane in this unique arid environment of Kuwait, which has not, as yet been addressed in research by the research communities. Therefore, the objectives of this research were as follows:

- To examine the behaviour of methane oxidising bacteria subjected to conditions of oxygen availability and sustainability
- To investigate the following conditions on methane oxidation in landfills existing in arid environments:
    - Suitability and effect of commonly available soil material in desert environment as a cover for the reduction of methane.
    - Effect of oxygen penetration level into a cover layer on methane oxidation.
- Based on the accumulated data obtained from the literature and from experimental tests, to propose a system of mitigation that could reduce methane production from landfills existing in arid climates, in preparation for future *in situ* field tests.
- To recommend a scheme and a course of action for the authorities in Kuwait to take, in order to reduce the carbon footprint resulting from landfill emissions.

Chapter II

Methanotrophs and Methane Oxidation Factors Affecting Oxidation in Landfills

2.1 Introduction

Methane gas production in landfills results from anaerobic microbial assimilation and degradation of organic matters under suitable temperatures, humidity, and other favourable conditions. However, a group of microorganisms, the methanotrophs, are able to oxidise methane gas under the right conditions and transform it into less harmful gas (carbonbe dioxide). These microscopic methanotrophs are germ-negative bacteria (Mancinelli, 1995) and are able to utilise methane as their sole source of carbon and energy. These methanotrophs were first identified by Sohngen (1906), but Wittenbury et al. (1970), then later by Hanson and Hanson (1996) that actual identification, classification, and characterisation of more than 100 organisms were done. Researchers and waste managers alike have considered these specific bacteria to be a possible means of mitigating landfill emission.

2.2 Methanotrophs and methane oxidation

Methanotrophs use unique enzymes, known as monooxygenases (MMO), to metabolise methane into methanol, using formaldehyde as an intermediate stage for its catalysis. MMO enzyme is the basic catalysing enzyme in methanotrophic bacteria and essential in methane metabolism. This enzyme takes two forms; namely, the particulate methane monooxygenases (pMMO) and the soluble monooxygenases (sMMO), both of which are present in all methanotrophs. However, it should be noted that the former can only be active in the presence of copper, when copper is available in certain level of concentrations; while the latter has a low dependence on copper (Hanson and Hanson, 1996). This uniqueness and the ability of methanotrophs to assimilate a large number of biotransformation are traits that have drawn researchers,

landfill engineers, and planners into utilising these microorganisms to control methane and their environmental contamination.

Methanotrophs that assimilate methane have been classified into three main categories such as Type I, Type II, and Type X, (Hanson and Hanson, 1996), as shown in Table 2.1. The first and third types use one pathway; while the second type uses another. These pathways take complex and detailed enzymes interaction inside the cells of the bacteria, as described in detail by Mancinelli (1995) and Hanson and Hanson (1996). These pathways are processes described by Hilger and Humer (2003) as in the following:

Ribulose monophosphate pathway (RuMP):

$$CH_4 + 1.5\ O_2 + 0.118\ NH_4^+ \rightarrow 0.118(C_4H_8O_2N) + 0.529\ CO_2 + 1.71\ H_2O + 0.118\ H^+ \quad (2.1)$$

The Serine pathway:

$$CH_4 + 1.57\ O_2 + 0.102\ NH_4^+ \rightarrow 0.102(C_4H_8O_2N) + 0.593\ CO_2 + 1.57\ H_2O + 0.102\ H^+ \quad (2.2)$$

Where ($C_4H_8O_2N$) is a biomass produced by the bacteria.

In these processes, through various reactions using cellular methane and MMO enzyme, energy is released by converting methane to methanol initially, and then to formaldehyde and other carbon compounds as intermediary products (Anthony, 1982; Hanson and Hanson, 1996). Figure 2.1 shows the interaction of these pathways, as described by Hanson and Hanson (1996).

It is worth noting that group X methanotrophs could utilise the ribulose monophosphate (RuMP) as primary pathway, and in the meantime, they possess low levels of other enzymes to utilise the serine pathway, and have a high growth rate at high temperatures, as dissimilarity features with group I.

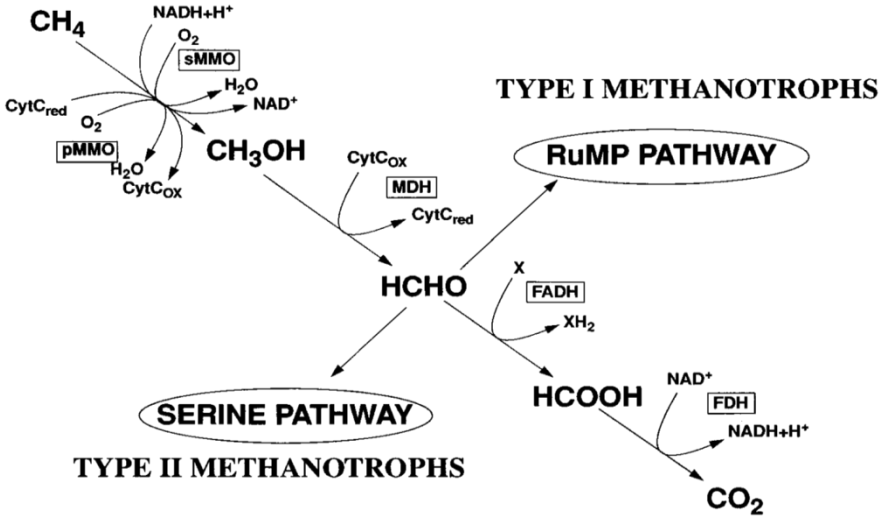

Figure 2.1 Interaction of pathways of methanotrophs and methane oxidation (source: Hanson and Hanson, 1996).

Methanotrophic bacteria are unique species in that, they are capable of utilising methane in a variety of environments and at different extreme conditions. In landfills, other microorganisms are important and present next to methanotrophs, and can contribute in consuming other non-methane landfill gases, some of which can affect the environment 1000-folds of greenhouse effects than methane (Scheutz et al., 2004; IPCC, 2007). This is worth considering when designing a system to control methane production, in order not to undermine their presence. Research showed that methanotrophs are present in sediments (Smith et al., 1997), groundwater (Filermans, et al., 1988), seawater (Holmes et al., 1995), peat bogs (Dedysh et al., 1998), hot springs (Bodrossy et al., 1997), salt storage (Khmelenina et al., 1996), and in the Antarctic (Bowman, et al., 1997). The concentration of bacteria can range from 10^3 to 10^6 cells per gram in the substrate of these environments (Hanson and Hanson, 1996). Therefore, the process of oxidising methane by methanotrophic bacteria in all of these environments could be considered as a carbon sink process. Table 2.1 shows some of the well-identified genera and their prominent morphological and physiological properties (Scheutz et al., 2009a).

Utilising these interesting features of the methanotrophic bacteria for the remediation of the environment and the earth, researchers should eventually focus their studies on

optimising the oxidation processes and on understanding their inner cellular works to build better systems for such remediation.

Genus	Phylogenetic position	ICM-arrangement (type)	Dominant PLFA	C-assimilation	Type of MMO	N_2-fixation
Methylobacter	γ-Proteobact.	stacks (I)	16:ω1	RuMP	pMMO	no
Methylocaldum	γ-Proteobact.	stacks (I)	16:ω1	RuMP	pMMO	No
Methylocapsa*	α-Proteobact.	Type III	18:ω1	Serine	pMMO	yes
Methylocella	α-Proteobact.	peripheral (II)	18:ω1	Serine	sMMO	Yes
Methylococcus	γ-Proteobact.	stacks (I)	16:ω1	RuMP	pMMO	Yes
Methylocystis	α-Proteobact.	peripheral (II)	18:ω1	Serine	sMMO /pMMO	Yes
Methylomicrobium	γ-Proteobact.	stacks (I)	16:ω1	RuMP	pMMO	No
Methylomonas	γ-Proteobact.	stacks (I)	16:ω1	RuMP	pMMO	No
Methylosinus	α-Proteobact.	peripheral (II)	18:ω1	Serine	sMMO /pMMO	Yes
Methylosphaera	γ-Proteobact.	stacks (I)	16:ω1	RuMP	pMMO	No
Methylothermus*	γ-Proteobact.	n. d.	n. d.	n. d.	pMMO	n. d.

ICM, intracytoplasmatic membranes; RuMP, ribulose monophosphate path; Serine, serine path; pMMO, particulate methance monooxygen – ase; sMMO, soluble methane monooxygenase; *genus represented by only one species; Proteobact., Proteobacteria; n. d., not determined, (PLFA), phospholipid fatty acid.

Table 2.1: Identified genera of methanotrophs and their properties (Source: Scheutz et al., 2009a)

2.3 Bulk gas composition and gas generation rates

Methane generation is an important feature of landfills because of its economic viability to the landfill industry. However, the estimation of the potential gas generated from these landfills is highly unpredictable, because of the many factors that affect the rate of decomposition. Waste composition, age of the waste, moisture content, waste distribution, level of acidity/alkalinity, nutrients availability, temperature, inhabitres, and many other factors, partially or combined could affect the rate of generation. In essence, each landfill has its own particularities, producing LFG dependent on the community that it serves. The primary component of landfill

gas is methane (CH_4), typically 40–55%, and carbon dioxide (CO_2), typically 35–50%; both are odourless gases and are site-specific. The typical bulk of landfill gases is specified by the US EPA (EPA-600/R-92-116) as described in Table 2.2, which includes considerable hydrocarbons, organic chlorine, and hydrogen sulfides compounds. These produced values by the US-EPA are specified as typical; however, methane and carbon dioxide, respectively, could be in the ranges of 40–70 and 30–60% (Shafi, et al., 2006); while the other contaminants in the gas could be found in the range of less than 0.5% (Brindley, 2012). Although these contaminants are small; albeit, they could pose a serious acute, repeatedly episodic, or prolonged hazard substances to humans (Brosseau and Heitz, 1994).

The hydrocarbons, chlorines, and sulfides contaminants have been counted by a study conducted by the UK Environmental Agency (EA, 2003) to contain 550 trace components present in the gas, regardless of whether the landfills were operated as municipal and hazardous collection sites (Eklund et al., 1998, Allen et al., 1997). Out of these components, the UK-EA have identified 25 landfill gas components to be toxic and carcinogenic, and published priority list to watch for, in and around landfills; these are produced in Table 2.3 (Shafi, et al., 2006).

Components	Typical landfill component level
Methane	40–55%
Carbon Dioxide	35–50%
Water Vapour	1–10%
Nitrogen	0–20%
Condensable Hydrocarbons	250–3000 ppmv
Organic Chlorine Compounds	30–300 µg/l
Hydrogen Sulfides	<200 ppm

Table 2.2: Typical landfills' bulk gas composition (US-EPA-600/R-92-116**)**

Priority trace component	Detection limit (mg m^{-3})	Component [x] (mg m^{-3})		
		[x] min	[x] max	[x] mean
1,1 –dichloroethane	0.02	<0.02	3.90	0.57
1,1 –dichloroethane	0.03	<0.03	19.0	2.24
1,2 –dichloroethane	0.07	0.13	46.0	5.71
1,3 –butadiene	0.02	<0.02	<0.02	<0.02
1-butanethiol	0.08	<0.08	<0.08	<0.08
1-pentene	0.16	0.24	21.0	5.49
1-propanethiol	0.04	<0.04	0.09	<0.05
2-butoxyethanol	0.04	<0.04	<0.05	<0.05
Arsenic	<0.01	<0.01	0.43	0.05
Benzene	0.03	3.10	73.0	18.4
Butyric acid	0.08	<0.08	17.5	1.85
Carbon disulphide	0.10	0.90	170	34.0
Chloroethene	0.02	<0.02	5.30	0.49
Chloroethene	0.30	1.10	730	102
Dimethyl disulphide	0.03	<0.03	12.0	1.02
Cimethyl sulphide	0.03	<0.03	24.3	3.69
Ethanal	0.01	0.08	2.55	0.43
Ethanethiol	0.08	<0.08	<0.08	<0.08
Ethyl butyrate	0.09	0.41	42.0	7.22
Furan	0.07	0.02	6.20	1.23
Hydrogen sulphide	0.15	2.40	580	111
Methanal	0.01	0.03	0.19	0.07
Methanethiol	0.30	<0.30	<0.30	<0.30
Tetrachloromethane	0.02	<0.02	<0.02	<0.02
Trichloroethene	0.04	0.25	88.0	8.59

Table 2.3: Priority list published by UK-EA (source: Shafi, et al., 2006)

Bulk gas production from landfill is not only important to the landfill industry, but also the rate of which it is produced in a yearly basis. Both the industry and the research community are trying to find ways and means to put estimates on the levels and rates of gas generated from landfills, where theoretical prediction models were used in order to determine these estimates. In these models, the particularities of each landfill, such as size, type of waste, moisture conditions, methane and carbon dioxide concentrations, soil type, hydraulic conductivities, etc., are measured and entered as input parameters into these models. Measuring techniques, such as soil core samplings, closed chamber, micrometeorological, mass balance, quantitative emission and trace measurements are techniques geared toward predicting gas and methane generation potential from biodegradable wastes. With this information as input, timeline-curves prediction series of the rates of gas to be collected from a

particular landfill are produced. An example of the outcome is illustrated in Figure 2.2, showing a typical prediction of methane production compared against the outcome of extracted actual data of gas collected for any typical landfill by time. While prediction and data typically show overall agreement; albeit, as expected, it did not generate a perfect match between them.

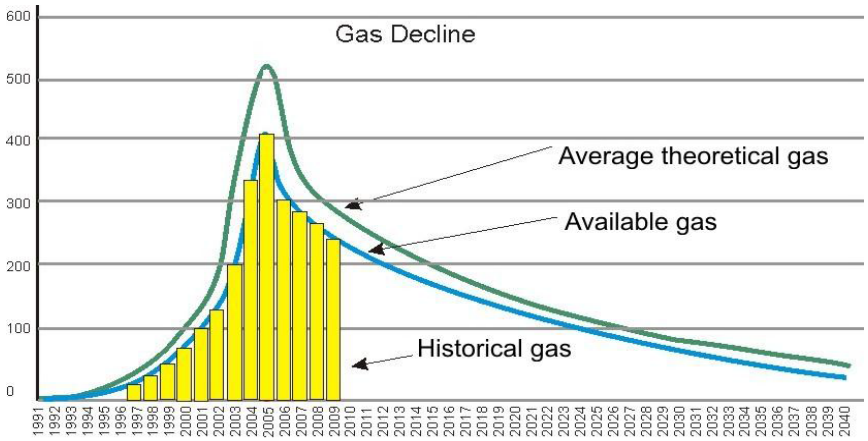

Figure 2.2: Comparison of theoretically predicted gas production curve with typical historical production data (source: Brindley, 2012).

Several models were developed to quantify the rate of gas generation rates annually (Fredenslund et al., 2007; Thompson et al., 2009; Oonk, 2010), driven by regulation reporting obligations, and by the need to quantify the methane generation rate for economic viability of the landfills. These mathematical models were developed on the basis of a first-order decay process, implying that landfill gas decay is gradual due to decomposition with the passing of time, taking into account two things, namely that the waste have been deposited for years apart, that there is high uncertainty of some input parameters, indicating that these models were just estimate models. In his review, Oonk (2010) estimated that these models can either over or underestimate gas generation rates by 25–50%; however, to avoid these over/underestimation outcomes, the curves outputs in the model (typical of Figure 2.2) should initially be validated and continuously updated, preferably on a yearly basis. Table 2.4 shows some of the freeware models that have been used extensively through the landfill industry for predicting the production of gases from MSW in landfills.

For overall gas production of a landfill, Ranson (2009) of the Worldwide Engineering, Environmental, Construction, and IT Services company in a report to the Ministry of Environment, British Columbia, USA, suggested that gas generation potential could reach to 20, 120, 160 m^3/ton of biodegradable wastes for relatively inert, moderately decomposable, and decomposable, respectively, for any given landfill. These potentials are for the lifetime of the landfills, and the decompositions are dependent, by a large extent, upon the annual rainfall water infiltrating the landfill cover, affecting anaerobic biodegradation.

Model	Country
IPCC-Model	Europe
TNO-Model	Dutch
GasSim	UK
LandGem	USA
Afvalzorg	Dutch
E-PRTR	Finland
Vogt-Model	California

Table 2.4: Freeware models for predicting the potential gas production rates for household MSW

To put it briefly, determining the rate and level of gas production from any particular landfill is to an extent, arbitrary and unpredictable and can only be estimated theoretically. These estimated outcomes must be verified and continually updated with the acquired actual landfill data during the operations.

2.4 Factors influencing methane oxidation in landfills

In adapting to its living environment, while methanotrophic bacteria are very versatile biological microorganisms; albeit, this versatility is influenced by the surrounding environments, which affect their own cell metabolism, and hence, their consumption of the surrounding substances engulfing them. These micro environmental conditions, in turn, are influenced by several prevailing factors that can inhibit the bacteria's own

activities and can influence the rate of methane consumption. Among others, adequate supply of oxygen and methane, right pH level, enough moisture content, right level of temperature, availability of nutrients, right soil conditions, and low level of inhibiting substance in the soil are some of the factors that are needed to be maintained for the proper functioning of bacteria, particularly in arid climates (Humer and Lechner, 1999). Therefore, to achieve the objective of identifying and quantifying the effects of these factors on the arid environment, and to attain the optimised engineering system for these climates, utilising the property of methane assimilation by the bacteria, it is imperative to conduct an in-depth review, study, and an understanding of these factors. It is also important to measure the influence of these factors on the rate of methane emission reduction. These factors and their characteristics in addition to the properties of the cover layer, that is the source term of methane production, are all discussed in details in Appendix E.

2.5 Formation of extra cellular polymeric substance (EPS)

Like most soil bacteria, methanotrophs are capable of producing EPS, comprising of sugars and amino acids (Hilger et al., 2000). This material has a high molecular weight, consisting mostly of polysaccharides produced in slime-like forms (Hou et al., 1978; Costerton et al., 1978) and has a function of anchoring the bacteria to the grains of the soil. EPS was observed to consist of 75–89% of polysaccharides and protein (Tsuneda et al., 2003). Bacteria also use EPS as protection against unfavourable conditions, such as high methane or oxygen loadings, predation, high or low heat conditions, and nutrients imbalance (Chiemchaisri et al., 2001; Fletcher, 1992; Babel, 1992). It was also observed that the bacteria of types I and II produce EPS material to prevent the accumulation of formaldehyde when the presence of carbon in the soil becomes high (Linton et al., 1986; Malashenko et al., 2001).

As was observed by Hilger et al. (2000), EPS production by methanotrophs is responsible for the gradual decrease in methane oxidation from peak values to lower steady values, as shown in Figure 2.3. This oxidation behaviour was reasoned to have been due either to the clogging action of pore spaces that are present in cover soil or to

the impeding of diffusion of gases. In the light of testing this behaviour, Hilger et al. (2000) performed a column test experiment, and confirmed that the data available support the idea that EPS can hinder oxygen diffusion into active oxidation layer and inhibit methane oxidation. This result implied clearly that EPS formation reduces methane oxidation in the soils. However, EPS formation was not of any significance, on the other hand, when field trials were carried out for a six-year duration of a passively vented bio-filter, made of porous clay pellets, even with high methane loading of 5929-g CH_4/ m^2/d (Gebert et al., 2006). This finding is consistent with the observations of Huber-Humer (2005) in a field trial of compost bio-covers, indicating that EPS production was insignificant, and that EPS can only form in hot spots even with high methane loadings. In contrast, observations from laboratory experiments proved otherwise. These findings on the effects of EPS on inhibiting methane oxidation could be attributed to the high aeration action in the open fields. In addition, when soils are composed of sizable aggregates and when rainwater washes away the EPS quickly as it forms, since EPS is a water-soluble material, that in addition to the continually changing ambient weather conditions, have contributed substantially to the EPS reduction in field tests. Moreover, lab experiments in contrast to the natural field settings bring high results due to the steady and continuous methane and oxygen loadings in confined spaces of the experimental apparatuses. These natural weather conditions, preventing the formation of EPS, are believed to be the reason behind the creation of hot spots observed occurring naturally in field experiments.

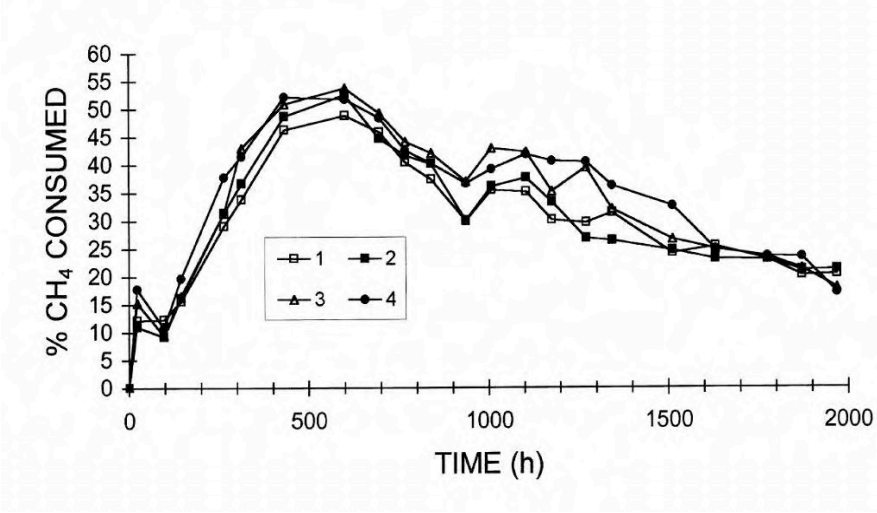

Figure 2.3: Reduction in methane oxidation of four soil column samples, due to EPS formation (source: Hilger et al., 2000).

2.6 Enhancing cover materials and cover technologies

Increasing the oxidation capacity in landfill covers requires a companion medium with optimum characteristics similar to the ones discussed in sections E.1–E.24 of Appendix E. Specifically, the medium must have the characteristics and quality of being coarse, porous, organically active, of favourable moisture contents, of appropriate temperature, etc., in order to create favourable environmental conditions for the methanotrophs. Materials used for the sole purpose of increasing the porosity of the soil for better diffusion of air were also experimented on. Such materials are crushed glass (Stem et al., 2007), gravel (Huber-Humer, 2004), plastics (Chiemchaisri et al., 2013), biochar (Reddy et al., 2014), and shredded tires (Park et al., 1996; Sadasivam et al., 2014). Some of these materials such as the plastics, when used as amendment or in distribution layers, may decompose, producing their own harmful gases and other leachate by-products, and others, such as glass, may not be suitable for the growth of the methanotrophs due to their smooth surfaces, hindering the attachment of the bacteria to the surface (Sections E.4 and E.5 of Appendix E).

Most laboratory and field tests results, when testing for enhanced cover materials, indicated that organic waste materials, such as composts, sewage sludge, peat, etc., used either solo or amended with other materials as additives, could mitigate methane effectively. The rate of methane removal, using these bio-based materials could reach up to 100%, given that the composted materials possess high porosity, high water holding capacity, and appropriate nutrient levels (Huber-Humer et al., 2009). In addition, the medium consisting of composts and the amendments should also have high permeability values for gas to pass easily. Likewise, they should have high aggregate sizes and fine texture to hold adequate gas at sufficient time to allow the bacteria to act on the conversion of methane (Stern et al., 2007). Furthermore, the medium should have the property to transport and hold volumes of air in its pore spaces even at high moisture contents, so that it can allow diffusion around these spaces to supply the bacteria with its needed oxygen. Finally, the compost material should have organic matter content in a biologically stable state, so that it cannot produce methane by its own making, thereafter consuming oxygen instead, depriving

the bacteria from their needed share of air. Nevertheless, all these desirable properties in composts come with huge challenges.

The maturity of this compost material is another major drawback when using composted materials. The process of maturing compost material, by itself, requires producing a large amount of methane, releasing it into the atmosphere as the material matures, then, moving the same material on to a landfill to capture methane in another setting. This process is actually harmful to the environment.

Producing a well-structured compost to be used on landfill covers requires a diverse process, including screening, adding straws at different times, controlling the temperature, moisture, etc. Different compost venders use different processes to produce composts, which make difficult their description and classification; since there is no classification system in place for compost material, sand, for example. In addition, since composts are seasonal and are not readily available everywhere and at any time throughout the year, it is not a viable option for landfill owners and operators. In developing countries, such as Kuwait, no production facilities exist, nor are there any importing agencies in the country to count on to make composts available as additive to landfill covers. Furthermore, there is the question of economic viability, considering costs in classification, transport, distribution on landfills, and construction. In particular, the costs will become even higher, if the recommendations in some studies were to be implemented, such as the use of a 120-cm thick layer of compost for high capacity of oxidation, covering the whole site (Huber-Humer, 2004; Stern et al., 2007). For these reasons, the advantages of using the bio-mitigation systems for landfill gas reduction as an alternative method to the conventional extraction or flaring techniques are thus, compromised.

Considering other technical difficulties, compost covers have the characteristic of shrinkage because of settlements in response to the alteration of wet and dry seasons. This shrinkage characteristic produces cracks and fissures in the cover system, producing with time, a preferred landfill gas path to flow, creating hot spots in the cover system and allowing methane gas to escape unoxidised (Czepiel et al., 1996; Chanton et al., 2011). This shrinkage also causes the material to compact up to 20%

(Scheutz et al., 2009b) as time evolves, reducing the porosity of the cover material. With that amount, the very characteristic advantage favouring the use of compost in the first place could result in the reduction of air diffusion and gas flux in the cover system. Moreover, when freshly placed, the compost material has the potential to allow water to seep through into the lower layers in fast movements, due to its high porosity, and thus, has the potential of creating more leachate pollution, affecting the underground water and its functionality.

Operationally, landfill cover made of composts has the possibility of allowing advective landfill gas to pass quickly through the layers, due to the pressure gradient inside the top cover and to the compost's high porosity, therefore, pushing oxygen in the pores and voids out of that cover layer. The process limits oxygen and air availability from diffusing into the cover system, hence, preventing atmospheric oxygen from reaching the bacteria. Additionally, the desired characteristics of high permeability and porosity of the compost material can increase the moisture content, reaching well over 30% w/w in the cover system in wet weathers, thereby, limiting oxygen penetration. Consequently, this condition tends to create an anaerobic condition, eventually, leading to the production of methane (Schuetz and Kjeldsen, 2004). Corollary to this, European Union (EU) countries and many other countries have required that only low permeability covers are to be installed on landfills (Eureopean Directive 1999/31/EC - Annex I, 3.1, EC, 1999) to be allowed operations. Such requirement may limit the use of compost-based cover systems on landfills. Also, laws and regulations have decreed stringent rules for waste handling and disposal, resulting in more research interests in optimisation and in disposal processes. As yet, there have been no regulatory institutions or municipal bodies that have endorsed, approved, or adopted the use of any new technologies involving compost materials, or low permeability materials. Thus far, none has ever been implemented on landfills. The difficulty of using compost materials with new measures stems from the fact that some uncontrollable and interlocking factors play major roles on the way gases are produced from landfills. Finally, arid environment that involved the precipitation of particulate matters have the potential to clog the surface of covers in the landfills, in which it could hinder air diffusion to the oxidation layers. Therefore, there is the need for continued research to

determine a more appropriate cover material or cover methane mitigating systems, at the very least with more advantages than the compost-based covers. In due course, further research would add to the existing knowledge toward optimisation efforts for better methane reduction.

Investigation of microbial methane oxidation systems on landfills has taken many directions, both in laboratory and field studies and is well- documented in the literature, among others (Whalen et al., 1990; Figueroa, 1993; Kightely et al., 1995; Boecks et al., 1996; Czepiel et al., 1996; Kjeldsen et al., 1997; Boerjesson et al., 1998; DeVisscher et al, 1999; Humer and Lechner, 2001; Scheutz and Kjeldsen, 2001; Huber-Humer, 2004; Gebert et al, 2005; Dever et al., 2007; Philopoulos et al., 2008; Einola et al., 2009; Bonger et al., 2010; Scheutz et al., 2011; Morris et al., 2012; Scheutz et al., 2013; Kjeldsen and Scheutz, 2014). However, the designs for methane oxidation systems are concentrated only on a set of proposed concepts, in which all researches revolved around the points of enhancing these concepts. Kjeldsen and Scheutz (2014) summarized these designed concept systems as follows: a full surface bio-cover, bio-window, open bed passive bio-filter, closed bed bio-filter, open bed, active bio-filter, closed bed, bio-filter, bioactive intercepting trench, and combined solutions. These systems and capture methods are discussed in detail in Appendix A.

Due to the difficulty in accounting for the exact level of methane generated from landfills, to include the costs involved in setting up *in situ* experiments in landfills, tests on different material amendments to simulate landfill covers system have taken two directions. The first is by using batch incubation, and the second is by the use of reactor columns. These test methods are an attempt to simulate the factors affecting oxidation in the actual setting of a landfill, in a more controlled setting of a laboratory and with lower costs involvement. As a whole, these testing methods can represent the circumstances existing in a landfill well, and an actual field-testing afterwards should ensue to augment their outcomes. The level of better soil representation of each one of these methods depends on the factors that are being tested. The following sections explain in detail, the advantages of each testing method and indicate through some examples, tested soils in relation to field tests.

2.7 Batch tests

Testing for methane oxidation capacity in a laboratory can be studied either by using an incubation chamber (batch test), or by setting up a column experiment. The batch tests are best used in experiments where large numbers of samples of bio-cover materials or conditions are investigated, as they offer the advantage of lower cost and simple experimental setups. This advantage is often utilised as a first step to compare various cover materials intended for use in a bio-cover system, prior to setting up subsequent column tests. In contrast, column experiments comprise a column (cylinder) often packed with layers of materials that best simulate the actual bio-cover system, with the advantage of allowing continuous flow and the precise specifics of environmental parameters. Material ranking, such as the sample batch data obtained from the literature, is technically easy and is an important step prior to embarking on a much costly column or field tests to select suitable cover materials. Table 2.5 shows recent batch studies, involving sands and compost materials for comparison, indicating that most compost tests produce higher oxidation rates over sandy loams. A high oxidation rate of 249-µg CH_4 g^{-1} h^{-1} has been obtained from a batch test, when a municipal solid waste (MSW) of 49% weight per dry weight (w/dw) organic matter content was used (Wilshusen et al., 2004a). Conversely, a low rate of 0.0024-µg CH_4 g^{-1} h^{-1} was obtained for sandy loam of 1.8% w/dw organic content (Boecks et al., 1996). However, data available from the literature are hard to compare, due to the different conditions used for each test, i.e., moisture contents, porosity, temperature, chemical contents of samples, etc. Furthermore, the units used in expressing the results are inconsistent. Nevertheless, the data would be helpful in showing indications of the capacities of the materials to oxidise methane.

The literature is full of other experimental batch testing (Schuetz et al., 2009), in which sandy soils are the most tested material with a mix of some type of organic matters. In these tests, organic matters of 2 to 5% w/w contents have been used with a methane concentration of more than 5% v/v; variables, such as temperature, moisture content, acidity level, inhibiting compounds, etc., modified in reactor chambers. High rates of

oxidation can be achieved in such systems. For example, methane oxidation rates of 644-μg CH_4 g^{-1} h^{-1} was possible when using a sandy loam sample, collected from the Coastal Marine Landfill, CA, USA, having 3.9% w/w organic matter, during tests at a 30°C (Spokas and Bogner, 2011).

Soil Material	Content of organic matter % w/dw	Moisture Content (MC) %(w/w)	Max. CH_4 Oxidation Rate	Temperature °C	References
Sandy Loam	1.8	15	0.0024 (μg CH_4 g^{-1} h^{-1})	25-30	Boecks and Van Cleemput, 1996
Sandy Loam	-	35	48 (μg CH_4 g^{-1} h^{-1})	38	Borjesson and Svensson, 1997
Sandy Loam	1-9	11-32	19 (μg CH_4 g^{-1} h^{-1})	15	Christophersen et al., 2000
MSW compost	49	123	249 (μg CH_4 g^{-1} h^{-1})	22	Wilshusen et al., 2004a
MSW (MBT) residue	47.4	49.5	28 (μg CH_4 g^{-1} h^{-1})	25	Mor et al., 2006
5-year old compost	7.3	21-28	2.5 (μg CH_4 g^{-1} h^{-1})	19	Einola et al., 2007
5-year old compost	-	10-15	16 (μg CH_4 g^{-1} h^{-1})	25-35	Park et al., 2009
Compost and wood chips (1:1)	14	35.1	5 (g CH_4 m^{-3} h^{-1})	22	Scheutz et al., 2009b
4 year garden waste compost	29	72	161 (μg CH_4 g^{-1} h^{-1})	Room	Pedersen et al., 2011
Sandy loam	-	75	14 (mg CH_4 Kg^{-1} h-1)	30	He et al., 2011
Mineralized refuse	55	55	14.73 (μmol g^{-1} h^{-1})	30	Zhang et al., 2012

Table 2.5: Summary of methane oxidation of selected data using batch tests

2.8 Column tests

Simulating landfill cover behaviour under laboratory conditions is best represented by a column test, where a vertical chamber (reactor) is built in a laboratory and packed with suitable material for testing. In this arrangement, input landfill gases can be injected in some suitable design scheme and environmental variables in the chamber adjusted continuously in order to test the reactor's capacity to oxidise methane. Such investigation can be costly and time consuming, particularly when various materials and conditions were in need of investigation; however, column reactor studies usually follow on from batch tests, and can very well represent circumstance of the field. An example of column test setups to study various environmental and design effects on methane oxidation in landfill covers have been reported by Scheutz et al., (2009b). A summary of recent results from column tests investigating methane is given in Table 2.6. Results have disclosed that a compost material, with or without additives, produces the highest rate of oxidation. The additives to the compost had been intended to enhance porosity of the mixture. A maximum rate of 583-µg CH_4 g^{-1} h^{-1} methane oxidation was observed for a system operated for approximately one year, under a loading of 589-µg CH_4 g^{-1} h^{-1}, achieving 100% rate of oxidation of methane at an ambient temperature of 19^{o}C (Haubrichs and Widmann, 2006). The same high rate of oxidation was achieved by Perdikea et al. (2007) for mature compost in a mix with small amounts of sawdust (9:1), with both studies agreeing with the results of Scheutz et al. (2009b), Huber-Humer et al. (2009), and Hrad et al. (2012). It is however interesting to note that methane oxidation outcomes using compost material are not always favourable. Schuetz et al. (2009) reported a steady state oxidation rate of -31-µg CH_4 g^{-1} h^{-1} for a compost residue, mixed with 50% sand under a load of 254-µg CH_4 g^{-1} h^{-1}, showing that methane was produced, instead of being assimilated; albeit, the organic material content was only 50%. This confirms that the use of mature compost material is essential, if it were to be used in any bio-cover system, and could explain the variation in methane achieved by other workers (Table 2.6). Likewise, sand particles, when mixed with compost, must have sufficient sizes to ensure an increase in the porosity of the mixture. The results shown in Table 2.6 should be taken only as an

indication, because variables and environmental conditions were not standardised in each set of experiments.

Soil Material	CH_4 Loading	Moisture Content (MC) % (w/w[a])	CH_4 Oxidation Rate Max (Efficiency)	Period (days)	Temperature (^oC)	References
Landfill sandy loam	368 (g CH_4 $m^{-2}d^{-1}$)	16.5	230 (g CH_4 $m^{-2}d^{-1}$) (79%)	65	Room	DeVisscher et al., 1999
Agricultural loam	216 (g CH_4 $m^{-2}d^{-1}$)	16.5	98(g CH_4 $m^{-2}d^{-1}$) (81%)	127		
One- year- old MSW compost	400(g CH_4 $m^{-2}d^{-1}$)	85	400(g CH_4 $m^{-2}d^{-1}$) (100%)	187	Room	Humer and Lechner, 2001
Loamy Sand	525(g CH_4 $m^{-2}d^{-1}$)	13	(83%)	30	Room	Park et al., 2002
Polystyrene pellets/Compost mix	200–500(g CH_4 $m^{-2}d^{-1}$)	--	242(g CH_4 $m^{-2}d^{-1}$) (72%)	86	Room	Powelson et al., 2006
Yard waste compost	589(g CH_4 $m^{-2}d^{-1}$)	32.2	583(g CH_4 $m^{-2}d^{-1}$) (100%)	369	19	Haubrichs and Widmann, 2006
Manure compost/Sawdust mix (9:1)	9.4 (g CH_4 m^{-2} h^{-1})	52	2-8 (g CH_4 m^{-2} h^{-1}) (100%)	40	22	Perdikea et al., 2007
Compost/Wood chips (1:1)	229-254 (g CH_4 m^{-2} d^{-1})	68	247 (g CH_4 $m^{-2}d^{-1}$) (58%)	255	22	Schuetz et al., 2009
Raw compost	198 (g CH_4 $m^{-2}d^{-1}$)	72	141 (g CH_4 $m^{-2}d^{-1}$) (55%)	111	Room	Pedersen et al., 2011
Compost/Soil mix (1:3)	-	22	420(g CH_4 $m^{-2}d^{-1}$)		25	Rose et al., 2012

[a] Dry weight.

Table 2.6: Selected oxidation data for bio-covers of landfills using column tests

2.9 Field tests

In order to determine the performance and capacity of a bio-cover to mitigate methane, and consequently, to obtain information for the design of a final bio-cover system, field experimentation comes as a final stage of testing after the preliminary laboratory tests. However, field testing presents challenges of controllability and quantifiability of the input and output parameters of the whole field site. Such variables are moisture content, permeability, diffusivity, conductivity, porosity, degree of compaction, as well as the rate of methane emission, amount of air (and oxygen) penetration, and nitrogen diffusion into the landfill surface. These difficulties explain the relatively low number of field tests conducted worldwide appearing in the literature (Kjeldesn and Schuetz, 2014). Table 2.7 shows some of the recent field trials conducted on existing landfills from different countries.

As discussed in Section E.8 (Appendix E), relative to results of several column tests, composted materials were found to be the most suitable materials to cause the highest rates of methane oxidation; therefore, they too were considered suitable for cover materials. Apparently, this could be the reason why investigators have been encouraged to use these composted materials as their primary materials for most of their field tests. Table 2.7 shows some of these field trials, indicating the efforts to use composted materials along with combination of soil and environmental factors to reach optimal oxidation levels. Cabral et al. (2010) obtained a high value of oxidation at 252-g CH_4 m^{-2} d^{-1} at high loading of 2212- g CH_4 m^{-2} d^{-1}, (Table 2.7), for industrial compost sludge in combination with sand mix (5:1). On the other hand, Schuetz et al. (2009) obtained 39- g CH_4 m^{-2} d^{-1} for methane loading of 39.5- g CH_4 m^{-2} d^{-1}, reaching to approximately 100% rate of oxidation, when using only municipal solid waste (MSW) compost. These results would infer that for high methane loadings, the oxidation rate is decreased in an inverse relationship with each other, confirming results shown in Figure E.8 (Appendix E). Another study conducted by Scheutz et al. (2011), using garden waste compost, produced only 41% rate of oxidation for a loading of 150- g CH_4 m^{-2} d^{-1}, showing different results from the MSW compost tried previously. This low oxidation rate could be attributed to the low porosity level present in the compost

material used, or to having used compost material that had not been matured or structured enough. The most interesting study in field trial, however, done recently in field trials, has been reported by Geck et al. (2013), who reported that top cover soil material can produce high oxidation levels, reaching as much as 96% for low methane load of 43- g CH_4 m^{-2} d^{-1}. This is indicative that materials, other than composted ones can practically produce high rates of oxidation, if the cover material and a cover system were to be favourable.

Considering all those aforementioned outcomes, results should be taken only as symptomatic, since each set of field trials is governed by its own pertinent soil and environmental conditions, and that some trials used different measuring methods from the others and with differing measuring units.

The use of non-consistent measuring techniques and methods in experimental and field tests has become a problem among researchers, as they have prevented the ability to compare directly results of their analysis. The usual field testing methods are carbon mass balance, stable carbon, mass flow, tracer techniques, flux chamber, and field flame ionisation methods. These different techniques used to measure methane are likely to cause discrepancies in their results. This prompted Huber-Humer et al. (2009) to dedicate a whole research article to address this problem, suggesting the use of a unified and a systematic approach, i.e., to use, for surface scanning, a combined flame-ionisation detector (FID) combined with chamber measurement. For larger-scale tests, a tracer technique would be most appropriate, which would require monitoring of the whole emission of a site, etc.

Units of measuring methane consumption for batch testing are done by using micro grams of oxidised methane per gram of soil sample per hour ($\mu gCH_4 g^{-1} h^{-1}$); while units of measurements of consumption in column and field tests are done by using a flux rate in grams of oxidised methane per square meter per day. These measuring techniques are dictated by the methods that are being used, since there are no fluxes involved in the batch experiments, in which to relate the results. The only means is the weight of the samples engulfed by gases, which have been used as a standard measurement for

batch tests. Similarly, there are no weight specifics of samples involved in the field tests, but only gases fluxing from the bottom of the field and air diffusing in flexuous fashion from the top. For column test, it is similar in arrangement to field tests, where methane and oxygen are introduced into the columns in a flexuous way; hence, the measurements are done in flexuous units. Therefore, these measuring methods are of different setups, and the measuring units are specific to each.

Soil Material	CH_4 Loading (g CH_4 $m^{-2}d^{-1}$)	CH_4 Oxidation Rate (efficiency)	Depth (m)	Methods for measuring CH_4 oxidation	References
Yard waste compost	54	36 (g CH_4 $m^{-2}d^{-1}$) (68%)	1.5	Carbon mass balance	Philopoulos et al., 2008
Yard waste compost/Wood chips (10%)	168	101(g CH_4 $m^{-2}d^{-1}$) (60%)	1.2	Carbon mass balance	Dever et al., 2007,2009
MSW compost/Wood chips (10%)	288	94(g CH_4 $m^{-2}d^{-1}$) (32%)	1.2	Carbon mass balance	
MSW compost	39.5	39(g CH_4 $m^{-2}d^{-1}$) (99%)		Reference cell	Huber-Humer et al., 2009
Compost Industrial sludge/Sand mix (5:1)	2212	252(g CH_4 $m^{-2}d^{-1}$) (11%)	0.9	Stable isotopes	Cabral et al., 2010
Fresh garden waste	300–700	(20–60%)	0.15	Flux chamber	Bongner et al., 2010
Garden waste compost	150	28(g CH_4 $m^{-2}d^{-1}$) (41%)	0.15	Flux chamber, Stable carbon isotope	Schuetz et al., 2011
Top soil	43	30(g CH_4 $m^{-2}d^{-1}$) (96%)	0.2	Flux chamber, Mass flow	Geck et al., 2013

Table 2.7: Selected field scale oxidation data of landfill cover studies

The Influence on methane elimination in the soil of a landfill is dictated by sets of factors. These main factors (shown in Table 2.8) can have direct effects on methane production, individually, or by having combined interlocking effects, which in turn, can play a major role in the production and mitigation of methane by the bacteria in the soil (Schenutz et al., 2009). Some of these factors are uncontrollable, and some are in contradiction with each other, in terms of reaching the optimal values, making it difficult for researchers to achieve an optimal engineered landfill system. This may be one of the major reasons why no study has ever claimed so far to have achieved an optimal barrier system for landfills. Not to discount some research efforts, thus far, only enhancements and optimisation of individual factors have been achieved successfully and have been the only means tried by researchers to reduce methane. The influence of some of these factors on methane elimination can be seen from the results shown in Figures E.7, E.8, E.10, E.11, E.12, E.13, E.15 (Appendix E), and Figure 4.23 for oxygen concentrations, methane loadings, internal gas pressures, moisture contents, NH_4^+ additives, temperature variations, pH variations, and organic matters amendment, respectively.

For engineers, waste planners, and researchers, these factors of interest are the ones that can be manipulated, managed, controlled, and constructed. Clearly, environmental and meteorological factors are not within reach of control, as well as the factors influencing landfill locations and ground characteristics, to some degree, in addition to waste characteristics factors. Site location and waste characteristics are somewhat more controllable than the environmental factors. Waste characteristic factors are influenced by the community and the presiding municipal authorities, which make site owners, city planners, and waste engineers unable to predict the characteristics of wastes deposited in landfills, even with the implementation of some collection and sorting regulations. The same applies to the site location and ground characteristics group of factors, thereby, leaving the soil and landfill cover characteristic factors as the only group that could be controlled, manipulated, and constructed. For the past decades, these sets of factors have been the subjects of research by researchers, producing varying and interesting results. Most of these factors in this

group are well-researched and well-understood by the scientific community, producing substantial information, except that a few still need further investigation.

Groups	Factors
Landfill location and ground characteristics	• Landfill latitude. • Landfill site waste capacity. • Site geological formation, presence of voids and cracks. • Site barriers. • Water table size and depth. • Landfill design and construction. • Vegetation and irrigation.
Waste characteristics	• Waste amount and composition. • Waste fermentation time. • Waste methane generation capacity. • Waste moisture contents. • Waste temperature.
Environmental and meteorological	• Environmental temperature. • Barometric pressure. • Wind turbulence and speed. • Rainfall, amount, and frequencies.
Soil and cover characteristics	• Cover layer thickness. • Compaction and density. • Soil granular types. • Porosity. • Water retention and holding capacity. • Moisture contents. • Hydraulic conductivity. • Thermal conductivity. • Heat conductivity. • Thermal capacity. • Diffusivity. • Permeability. • Soil acidity. • Nutrients and minerals. • Inhibiting substances.

Table 2.8: Summary of factors affecting methane emission

One of the factors that needs further investigation is oxygen delivery through the cover surface and its sustainability inside the cover layers, if the cover surface is controlled by

regulation or if clogged by dust, in the case of arid environment. As seen from Figure E.6 (Appendix E), oxygen availability in the top cover at its best was to reach a level of 10–20 cm from the top that could support methanotrophic bacteria and no more than that depth level if the other conditions were not favourable, i.e., high-pressure gradient in soil, high moisture contents, low soil porosity, etc., would prevent oxygen from diffusing into the soil, even to lesser depth than that of the desired level. Schuetz et al. (2004) concluded that the capacity of bio-based systems to oxidise methane is related to the depth of oxygen penetration, and that the maximum oxidation that can occur is at depths of 15–20 cm, below the surface of the soil, as data have suggested from field experiment conducted on Skellingsted landfill, south of Holbaek, Denmark. So concluded, Barlaz et al. (2004) confirmed that the extent of oxygen availability limits methane oxidation, and that, the cover design that enhances oxygen availability could enhance methane oxidation. Several other researchers have also arrived at the same conclusion (Molins et al., 2008; Scheutz et al., 2009b; Gebert and Gorengroeft, 2009; Cao and Staszewska, 2011). Thus, it is important to address this limitation of oxygen availability when setting out to introduce a system of bio-cover to mitigate landfill methane in arid environment.

2.10 Optimum factors in relation landfills located in arid zones

Reviewing and analysing the factors responsible for methane oxidation in landfills, as discussed in detail (Sections E.1-E.24 in Appendix E)), were deemed necessary in order to understand the effects of these factors on methanotrophic activities in methane reduction in landfills, and because these factors are interdependent. In addition, it is also necessary to understand the limitation and capacity of the bacteria to assimilate methane for methane reduction, and finally, to realize and understand the optimal ranges of these factors. This task of finding an optimum practical barrier for arid environment, however, is obviously considerable. Focusing the research on specific element/factors could enhance the understanding of landfill emission in arid zones.

Most factors/elements reviewed and analysed previously, showed some optimal limits or favoured ranges by which assimilation of methane can be done efficiently by the

bacteria; however, optimal limits of some other factors could not be realised due to the nature of the factors themselves, or due to the limited research done on their respective areas. For example, phosphate nutrient optimal values are not available; their effects on oxidation are not clear, and optimal wind turbulence effects are also not easily obtainable, being with limited available data. Other factors, on the other hand have the nature of inhibiting oxidation rather than enhancing it, i.e., the formation of EPSs. A summary of available optimal factors/elements affecting methanotrophic bacteria's activities and the oxidation process confirmed in the literature are shown in Table 2.9.

Obtaining an optimum and practical system to reduce landfill emission in arid climates by utilising microbial oxidation requires assessing these factors fully. The aforementioned analysed factors and particularly the ones summarized in Table 2.9 encompass the most effective elements that directly influence the conversion of methane by methanotrophic bacteria in landfills to CO_2 and other products. Some of these factors however, have a conflicting nature. For example, the more the element of porosity, which allows the filtration and diffusion of air into the landfill cover soil is allowed, the more likely that moisture, leachate, and soil particles seep into the soil, hence, affecting the process of methane production or its elimination. This is more seen in desert environment, where frequent sandstorms cover the surface quite often. This conflicting effect can either be by the individual element, or by having a combined synergistic effect of it and the other factors. Maintaining porosity even at optimum values as indicated in Table 2.9 does not guarantee a favourable condition for the methanotrophs to be fully active in the soil. An increase in porosity means an increase in particle sizes, allowing more spaces and voids in the cover layer. This favourable setup decreases the specific surface area of the soil; hence, higher diffusion of higher porosity is met with low specific surface area at the same time. Also, the more nutrients there are in the cover soil via fresh additives (i.e., added ammonia, premature compost, etc.) the more likely oxidation of methane would be enhanced, and also stimulate other bacterial growth, which would allow competition for oxygen. Such combined interdependent interactions exist for many of the factors shown in Table 2.9, illustrating the complex nature of the landfill cover systems.

Elements / Factors	Optimal range	References
Landfill cover characteristics		
a) Cover Layer	120 cm 40 – 50 cm	Huber – Humer, 2004 Martikkala and Kettunen, 2003
b) Compaction density	0.8-1.1 ton/m^3	Huber – Humer et al. ,2009
c) Soil grain sizes	50 µm–2mm Coa rse grain sizes	Bender and Conrad, 1995 Huber – Humer, 2004 Huber – Humer et al., 2008, 2009 Scheutz et al., 2008
d) Water retention and holding capacity	50–130% of Dry matter (DM)	Huber – Humer et al., 2009
e) Hydraulic conductivity of municipal waste	$5.9x10^{-7}$–$2.6x10^{-2}$m/s	Landva and Clark, 1986,; Ettala, 1987;, Oweis et al., 1990,; Shank, 1993,; Jain et al., 2006
Oxygen supply	21% (v/v), Type I 1% (v/v), Type II	Herckel et al.,,2000 Erwin et al., 2005
Methane loadings	60g $CH_4m^{-3}h^{-1}$ Compost bio-filter	Streece and Stegman, 2003
Moisture content	13–15.5% (wt/wt) Upper layers 25–50% (wt/wt) Compost material 15–30% (wt/wt) Soil cover	Httiaratchi, 2001 Humer and Lechner, 1999 Scheutz and Kjeldsen, 2004
Nutrients	25-mg N/kg soil NH_4, NO_3 1-5 µ Mol/L,Cu 0.020 -kg/kg soil, Cu 0.1–5.0 mg/L, Fr	Hettiarachi et al., 2000 Hanson and Hanson, 1996 Mohanty et al., 2000 Boiesen et al., 1993
Temperature	20–30°C 25–35°C 20–30°C 30°C 18°C	Boeckx et al., 1996 Borjessen and Stevensson, 1997 Nesbit, 1992 Whalen et al., 1990 Humen and Lechner, 2001
PH-acidity	5.5–8.5 6.7–7.5 6.6–6.8	Scheutz and Kjeldsen, 2004 Bender and Cornard, 1995 Whittenbury et al., 1970

Table 2.9: Optimal values of elements/factors affecting methane oxidation confirmed in the literature

Factors/elements discussed in Sections E.1 through E.24 (in Appendix E) may not be the only elements to have direct effects on landfill methane assimilation, reduction, or released into the environment. Other physical factors could also have greater effects. The type and design of the physical containments involve alteration to the cover soil, therefore affecting the activities of the methanotrophs, as these top cover containments are most sensitive to disturbances, as seen from previous discussions. These designs have been investigated thoroughly in the literature. These cover designs, materials, and arrangements are presented in Appendix A.

Table 2.9 shows some of the more important factors/elements that have direct effect on methane assimilation in landfill cover, and when compared with measured factors existing in arid landfill, the results are surprisingly almost similar. Table 2.10 shows some of landfill measurement attributes based on Al-Qurain landfill measurement in Kuwait. Temperature ranged between 23 and 35°C inside the landfill, even though the ambient temperature could reach up to 48°C, which indicates high insulation properties of the soil existing at the landfill. In addition, the high liquid wastes deposited along with high volumes of organic matters, bring moisture content of the waste to a range of 15–40% (wt/wt), and when evaporating through the surface, it could bring cooling to the landfill, and hence, could reduce the temperatures. Temperature and moisture content ranges are close to optimum values; however, pH values, that range between 7.8 and 8.0 are a little alkaline, although within the optimum range presented by Scheutz and Kjeldsen (2004). Soil conductivity is another variable which is different from waste conductivity, where the need for both soil cover and waste conductivities, should be of high levels, for oxygen and methane diffusion. However, the soil conductivity in Al-Qurain landfill is very low for oxygen diffusion. Other measurements and attributes for the Al-Qurain landfill are not available.

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Factor	Average Level	Reference
Temperature	23–35°C	Al-Yagout et al., 2005
Soil hydraulic conductivity	4.0 x 10^{-7} m/s	Al-Yagout and Townsend, 2004
pH	7.82–8.06	Al-Yagout et al., 2005
Moisture content	15–40%	Al-Yagout, et al., 2007
LFG production	312+/- 125.2 ml/min	Al-Yagout et al., 2005

Table 2.10: Typical attribute measurements of Al-Qurain landfill in Kuwait

Chapter III
Experimental Investigations: Methodologies

3.1 Introduction

Kuwait soil is a hardpan calcareous soil, a sandy matrix with high silica content, with slight gypsiferous mix (Yamane, 1970), with characteristics summarized in Table 3.1, and chemical makeup presented in Table 3.2. The soil is mostly coarse sandy soil at higher depths (loamy sand) and almost equal in granular makeup between coarse and fine sands on the surface (sandy soil). The soil is slightly acidic and poor in nitrogen in deeper layers, with potassium, sodium, and chloride salts, with very high sulfates. This countrywide cover material must be dealt with when placing a cover over any landfill. This material has high fine sands and high pH makeup, making it unfavourable for landfill use, due in part to its low hydraulic conductivity (4.0×10^{-7} cm/s) to allow the diffusion of oxygen to the inside of the top cover. Table 2.10 shows the measured landfill attributes for one of the closed landfills in Kuwait, showing favourable conditions for anaerobic methanogenic activities, but less suitability for aerobic methanotrophic methane assimilation. The treatment for such highly impermeable soils to allow better oxygen diffusion for landfills is to use one of the new technologies being suggested lately (Appendix A). However, given the high dust fallout in arid climates, which is more persistent in Kuwait than most countries, it is unlikely that any one of these new technologies would be applicable. The dust fallout, described in Section 1.3, deposits fine particles that can easily penetrate and clog the surface pores of any cover system placed on any landfill. Therefore, to solve this diffusion problem, the air (and oxygen) diffusion into the landfill surface must be dealt with differently in this unique environmental condition.

Depth (cm)	Coarse sand 2–0.2 mm	Fine sand 0.2–0.02 mm	Silt 0.02–0.002 mm	Clay <0.002 mm	Classification
0.0 – 0.5	41.13%	48.0%	4.5%	6.2%	Sandy soil
0.5 – 13	46.6	39.8	7.4	6.2	
13 – 26	54.3	29.1	9.8	6.8	
26 – 40	62.5	24.5	7.6	5.4	
40 –	56.9	25.8	11.7	5.6	Loamy sand

Table 3.1: Kuwait soil's granular characteristics (source: Yamane, 1970)

Depth (cm)	pH	N (%)	PO_5 (mg/100 g)	Na_2O (mg/100 g)	K_2O (mg/100 g)	Cl (mg/100 g)	$SO_4^{-}S$ (mg/100 g)
0.0–0.5	7.50	0.011	6.2	6.0	5.0	1.8	236
0.5 –13	7.53	0.005	2.5	4.0	4.5	0.6	264
13 – 26	7.45	0.010	3.8	18.5	7.5	8.6	272
26 – 40	7.32	0.006	4.6	92.5	15.5	41.0	295
40 –	7.45	0.005	3.7	162	16.5	82.5	74.7

Table 3.2: Kuwait soil's chemical characteristics (source: Yamane, 1970)

Following the objectives identified in Chapter I, both batch and column tests were used. For the effect of oxygen delivery and sustainability on methanotrophic bacteria's activities, a batch test was used, and column tests ensued thereafter to further investigate oxygen delivery and the sustainability for commonly available materials in arid environment, and on the assumption that the column tests had the flexibility to test several variables simultaneously. These procedures are described in more detail in this chapter.

To fine-tune the objectives of this research, to reach the final aim of proposing a more efficient cover system for mitigating methane produced from landfills operating in arid climates, the steps taken were the following:

- Investigation of soil samples obtained from the top cover of a locally managed landfill, to obtain their oxidation profiles.
- Investigation of these soils samples under shaking and static conditions to observe the methane oxidation profiles.
- Investigation of the oxidation time lag of the samples that were taken from different methane exposed locations of the local landfill subjected to methane and oxygen loads.
- Comparison of the oxidation profile of these landfill samples with freshly taken soil samples obtained from normal common (university) grounds.
- investigation of the effects of oxygen availability on the oxidation rates of all sampled soils.

- Investigation of the effects of oxygen supply on the oxidation profile of tried samples, when subjected to intermittent oxygen loadings.
- To set up a column reactor experiment to test for commonly available materials, suitable for arid environment, to obtain a methane oxidation profile.
- To set up another column reactor experiment to test for the effect of oxygen delivery deep inside and within the soil for the mitigation of methane, and to compare with conventionally atmospheric driven delivery systems.
- Based on the information obtained from the literature and from the results of experiments, a practical solution was to be proposed for better oxygen diffusion into soil, suitable for arid environment.

3.2 Oxygen availability and dependence for sustained methanotrophic activity

The presence of oxygen in the microenvironment of the bacteria is a factor of utmost significance, in that the bacterium needs to metabolise methane to sustain life. In this process, to produce CO_2 and other by-products (as discussed in Chapter II), it needs oxygen as a reactive agent. This fact has motivated researchers to search for optimised landfill materials that are environmental- friendly, readily available, and cost-effective for oxygen availability to the bacteria. All efforts have yielded mixed outcomes, with only one notable material that stood out, i.e., the compost material as a landfill amendment. However, this too has its own drawbacks, as discussed in Sections 2.6–2.9.

As mentioned in Sections E.5, E.9, and E.10 (Appendix E), several researchers have investigated the availability of oxygen and its delivery to landfill covers. In addition, the time for the methanotrophs to regenerate and start digesting methane in landfills is critical, as this can affect the amount of pollutants emitted into the atmosphere. It is therefore vital to investigate these factors using methane-degrading bacteria obtained from locally selected landfill soil environments, in addition to asserting the importance of oxygen availability in the soil, and more importantly, its sustainability. With these two objectives in mind at this stage of the investigation, an experimental batch setup was arranged.

3.3 Batch tests

The methodology to verify the two aforementioned sub-objectives, namely, oxygen availability and its sustainability, a batch experiment was set up. The purpose was to investigate the behaviour of the methanotrophic bacteria that have been exposed to intermittent methane and to test their capabilities of oxidation when placed in different types of soils.

3.3.1 *Methanotrophic activity in different soil types*

Samples from locally closed landfill and other places were collected by scooping several grams of top surface soil, placed in sealed containers and then brought to the laboratory. These samples and leachates were taken from low and highly exposed methane locations; their makeup is described in Table 3.3. Unfortunately, back history of methane exposure of these samples was not available. Establishing a new history database of exposure would entail lengthy time and investigation, requiring setting up specific equipment and landfill entrance permissions, which were neither available/nor possible. Nevertheless, some of the samples have been taken from top cover of methane emitting landfill and exposed to methane gas, and can be used as an indicative sample simulating exposed landfill cover (Sections 2.2, 2.3, E.12).

For this batch experiment, the supplies and equipment used were as follows:

- Wheaton glass serum bottle (volume 125 ml, clear, Cat. Z114014-12EA),
- Butyl rubber stoppers (gray, size 20 mm, Cat. 27232), and
- Aluminium crimp seal (silver aluminium removable centre, diam. 20 mm, Cat. 27227-U).
- SGE Gas Tight Syringe (with interchangeable needle, volume 100 µL, needle size 25 ga (bevel tip), 100R-V-GT) was used,
- Methane gas standards ((1%, 10%, 30%, 70%), with NITROGEN BALANCE, 112 L from Scientific and Technical Gases, Ltd),
- Septa for the GC (Molded Thermogreen LB-2 Septa, solid discs diam. 11 mm),

- A Gas Chromatography analysis (GC) with Flame Ionisation Detector (FID) was performed on a Carlo Erba HRGC 5160 mega series GC.

Sample Material	Soil Granular Properties	Soil Classification	Description	GPS Locations
Coxhoe landfill soil sample (Land-GCP)	2.0–5mm 9.1% 0.2–2.0mm 19.6% 0.063-0.2mm 24.7% <0.063mm 46.6%	Sand and silt	Taken from near top gas collection pipe	
Coxhoe landfill soil sample (Land-TLS)	2.0–5mm 5.2% 0.2–2.0mm 21.1% 0.063–0.2mm 26.5% <0.063mm 47.2%	Sand and silt	Taken from top lower side of the landfill	54°43'16.8"N 1°29'20.2"W
Coxhoe landfill soil sample (Land-MCP)	2.0-5 mm 3.5% 0.2–2.0 mm 18.5% 0.063–0.2 mm 28.4% <0.063 mm 49.6%	Sand and silt	Taken from near methane collection pipe	
Coxhoe landfill soil sample (Land-LCP)	2.0–5 mm 8.0% 0.2–2.0 mm 20.3% 0.063–0.2 mm 27.0% <0.063 mm 44.7%	Sand and silt	Taken from near leachate collection pipe	
River Tyne soil sample (River-BRT)	2.0–5 mm 0.15% 0.2–2.0 mm 0.92% 0.063–0.2 mm 24.4% <0.063 mm 74.53% (mostly sand and silt)	Sand and silt	Taken from the bank of river Tyne, Newcastle	54°58'16.1"N 1°42'11.5"W
Common ground soil (Com-GS)	2.0–5mm 5% 0.2–2.0mm 52.2% 0.063–0.2mm 25.0% <0.063mm 17.8%	Sand	Taken from the University of Newcastle's grounds, outside Cassie building	54°58'51.3"N 1°36'54.6"W
Pure methanotrophs culture (Pure)	Liquid	Culture	At the care of Dr. Angela of the Department of Civil Engineering and Geosciences, taken by permission	

Table 3.3: Description of sample used in the batch tests

Several 1000-ml and 160-ml bottles were first prepared by sterilising, and disinfecting, using autoclave apparatus. The different landfill soils and leachates samples, that were taken from Coxhoe Landfill[1], were all measured, sifted with 2-mm sized sieve, divided, and a 20-ml portion was extracted from each of these samples, and placed immediately in the sterilised bottles. These new divided samples are listed in Table 3.4. Nutrients media solution of 10 ml (Appendix B) was mixed with the landfill sample and with other samples collected from other places, as described in Table 3.4, shaken to make them consistent and uniform, flushed with continuous air for few minutes and then capped. The remaining volume space of the reactors was filled with 30% methane and 70% of standard room air (21% oxygen in air), which was achieved by drawing air out, first using a syringe, and then injecting the desired methane amount. All reactors were left to incubate at the same room temperature (22+-2^{o}C) and light conditions in the laboratory. In addition, duplicate blank (control) reactors were flushed with air, capped, 30% methane was introduced in each, and placed alongside the rest of the reactors. Methane percentage content in each of the incubators was measured continuously by drawing 100 µl of the gas sample from each daily using the gas tight syringe for over one-month duration and analysed. The measurement started after one day of incubation, using the GC analyser with FID. This kind of measurement was used as a way of measuring methane consumption. Methane consumption was calculated on the basis of volume change, using the formula as follows:

$$\% \text{ methane consumption (efficiency) (v/v)} = (V_{initial} - V_{final} / V_{initial}) \times 100 \quad (3.1)$$

The syringe used to draw gases from the head space is 100-µL SGE gastight syringes pre-fitted with removable needle and valve (100R-V-GT). The GC separation was performed on a capillary HP-Plot-Q phase column (30 m x 0.320 mm i.d) coated with 20-µm film thickness (Agilent Technologies, Palo Alto, USA). The injection port used a split ratio of 10 and was heated to 200^{o}C.

[1] Landfill Site, Coxhoe, Durham, DH6 4RT

The GC was held isothermally at 36°C, with hydrogen as the carrier gas (flow rate of 30 ml min^{-1}, initial pressure 55 kPa). Instrumental quantification was calibrated using standard methane gas (Scientific and Technical Gases, Staffordshire, UK).

Sample	Sample size.	Soil type	Added solution
Land-GCP (a)	20 ml	Landfill top soil samples in 160-ml bottles, from different locations (taken from Coxhoe landfill, Newcastle, UK)	10 ml of Media solution (Appendix B)
Land-GCP (b)			
Land-TLS (a)			
Land-TLS (b)			
Land-MCP (a)			
Land-MCP (b)			
Land-LCP (a)		Landfill top soil with leachate sample in 1000-ml bottles	
Land-LCP (b)			
River-BRT		River Tyne (UK) sample in 160-ml bottle	
Pure (a)		Newcastle University pure culture in 160-ml bottle*	
Pure (b)			
Land-L-Active		leachate sample 1 in 1000-ml bottles, active landfill	
Land-l-Old		leachate sample 2 in 1000-ml bottles, old landfill	
Land-Mix		Leachate Sample mix of Land- Active and Land-Old in 1000- ml bottle	

Table 3.4: Landfill soil samples (in duplicates, a, b) with added media solution

For quality control and assurance, duplicate control reactor, filled with standard air and methane in the same ratio of 70:30%, respectively, as the set of the experimental reactors, capped, and placed on bench with the rest of the samples. All samples of the experiments were conducted in duplicates and sampled in duplicates, as well as calibrating the instruments for every use with a minimum of five-point calibration in duplicates (10 points calibration curve). A sample of accuracy curve is shown in Figure 3.1.

3.3.2 *Soil samples diluted with media solution*

Since the first set of experiment did not produce appreciable methanotrophic reaction, and also to have the samples exposed more to the mixture of gases in the reactors, another set of samples with smaller portion of the same soil samples and with more added nutrient media solution was prepared, as shown in Table 3.5. The samples were again placed in another sterilised batch reactors and subjected to the same conditions as in the previous set, except that only 1 ml of collected subculture samples was mixed with 10 ml of nutrients media solution. The same measurement procedures were also taken in this set of experiment, as was done previously.

Sample	Sample size	Soil type	Added solution
Land-GCP (a)	1 ml	Landfill top soil samples in 160-ml bottles, from different locations (taken from Coxhoe landfill, Newcastle, UK)	10 ml of Media solution
Land-GCP (b)			
Land-TLS (a)			
Land-TLS (b)			
Land-MCP (a)			
Land-MCP (b)			
Land-LCP (a)		Landfill top soil with leachate sample in 1000-ml bottles	
Land-LCP (b)			
River-BRT		River Tyne (UK) sample in 160-ml bottle	
Pure (a)		Newcastle University pure culture in 160-ml bottle*	
Pure (b)			
Land-L-Active		leachate sample 1 in 1000-ml bottles, active landfill	
Land-L-Old		leachate sample 2 in 1000-ml bottles, old landfill	
Land-Mix		Leachate Sample mix of Land- Active and Land-Old in 1000- ml bottle	

Table 3.5: Soil subculture samples in duplicates (a, b) with Increased media solution.

3.3.3 *Bacterial behaviour in differently exposed soils to methane*

To investigate the behaviour of the bacteria that is present in soils that were exposed to high methane loadings compared to low exposure, a third set of samples were

collected from different location sites in the landfill; some were collected near the methane collection pipes; some were collected away from the pipes. Besides these samples, another set of samples was also collected from the campus grounds at Newcastle University, which had little or no methane exposure. Table 3.6 shows these collected soil samples.

Batch reactors tests of these samples in duplicates were again organized and implemented to ensure accuracy, introducing approximately 22% v/v methane in each batch test; while the rest was filled with room air, and all other conditions were kept the same as the previous set of samples.

Sample	Soil type	Added media
Land-GCP (a)	Near top gas collection pipe	One gram of sample added into 10 ml of media solution
Land-GCP (b)		
Land-TLS (a)	Landfill lower side	
Land-TLS (b)		
Land-MCP (a)	Near methane collection pipe	
Land-MCP (b)		
Land-LCP (a)	Near Leachate collection pipe	
Land-LCP (b)		
Com-GS (a)	Garden soil	
Com-GS (b)		

Table 3.6: Low and highly exposed methane soils samples in duplicates (a, b).

3.3.4 *Bacterial response to oxygen availability*

The effect of oxygen concentration on the methanotrophic activities in the soil is well-known and well-documented in the literature; however, the presence of oxygen and its

penetration into the soil of various types are in need of further investigation. When oxygen is diffused via the diffusion mechanisms discussed in Section E.10 (Appendix E), the gas attempts to overcome obstacles within the soil's microstructure to reach the methanogenic bacteria colonies present in the lower layers of the soil. For this set of experiments, the reaction of methanotrophs in the soil samples that were collected previously, in relation to oxygen availability, should be further understood. Therefore, another set of soil samples was obtained from a garden at the University of Newcastle campus grounds. The soil samples were mixed with sand, distilled water, and nutrients, and were prepared in duplicates similar to the previous samples for batch reactors investigation. These sets were split into two groups of the same composition as such: one was incubated statically at room temperature; while the other was mixed continuously on a shaking platform at the same room temperature. The shaking condition was intended to enhance oxygen diffusion into the samples. These two groups of samples were compared with the earlier samples that were collected from landfill locations taken from specific points on the local landfill, as described in Table 3.5, in order to gauge the methanotrophic reaction to the oxygen and methane presence. All of these were tested under the same set of constants of temperature, moisture, and nutrients, as that of all of the previous sets of tests in Section 3.3.1. These new sets of samples are described in Table 3.7. In each of these sets, methane was introduced as a percentage of volume per volume by replacing air volume with the same volume of methane using a syringe, as described in Section 3.3.1. Methane rate of consumption was calculated in accordance to equation 3.1.

In addition to the standard air volume existing in the reactor chambers (21% oxygen), an injection of oxygen was also introduced into all of the samples in this set of experiment using a syringe. Ten-milliliter volume of oxygen was introduced at two separate times for all of the samples described in Table 3.7 at the time when it was observed that the rate of oxidation has decreased. The reason for introducing oxygen at these two different times was to understand the effects of oxygen availability on oxidation, and its effect on the bacteria if it were to be made available on an intermittent or continuous presence, besides the existing static oxygen in the chambers. The batch reactors containing the samples, subjected to shaking condition,

were intended to allow oxygen and nutrients to mix and oxygen to penetrate further into the body of the samples.

Sample No.	Status	Soil Mix.
Com-GS-SHK (a)	Placed on shaking platform	University grounds garden soil Sample
Com-GS-SHK (b)		
Com-SD-SHK (a)		University grounds garden soil with sand
Com-SD-SHK (b)		
Com-DW-SHK (a)		University grounds garden with distilled water
Com-DW-SHK (b)		
Com-NT-SHK (a)		University grounds garden with nutrients
Com-NT-SHK (b)		
Com-GS-STL (a)	Placed static on bench	University grounds garden soil sample
Com-GS-STL (b)		
Com-SD-STL (a)		University grounds garden soil with sand
Com-SD-STL (b)		
Com-DW-STL (a)		University grounds garden soil with distilled water
Com-DW-STL (b)		
Com-NT-STL (a)		University grounds garden soil with nutrients
Com-NT-STL (b)		
Landfill Sample 1, Near top gas collection pipe. (Land-GCP-SHK)	Placed on shaking platform	1-ml subculture in 10-ml nutrients media solution
Landfill Sample 2, Lower side location. (land-TLS-SHK)		
Landfill Sample 3, Near methane collection pipe.(Land-MCP-SHK)		
Landfill Sample 4, Near leachate collection pipe. (Land-LCP-SHK)		
River Tyne sample, taken from riverbank. (River-BRT-SHK)		

Table 3.7: Comparison of soil sample types and conditions (in duplicates, a, b).

3.4 Column tests

Column testing is a second step closer toward field tests, which has the advantages of providing continuous flow of gases over a bed of soil materials and of altering variables

readily for better investigation results. In the second phase of this study, tests were set up to investigate two sets of parameters using the column experiments. The first was to test for the oxidation profile of sand and soil materials, simulating common materials in arid environment, and the second was to test for the effect of providing air deep inside the soil on the methane elimination from a landfill soil.

3.4.1 *Column test for common materials*

A material that had been widely experimented with and accepted as the most promising material for landfill cover was compost, as seen in Section 2.6. The logic behind this acceptance was that this material has been observed to possess some preferable characteristics of permeability, porosity, high organic matter, grain/void ratio, and high bacterial population, which in turn, had led to high oxidation rates. Nevertheless, despite these features, there were serious disadvantages to be considered. Among others, it is an alien and superficial material that is not commonly available, as evident from the discussion in Section 2.6. Therefore, to discard some of these drawbacks, materials like sand and soils that are more common, abundant, and readily available as cover materials everywhere, particularly in some arid countries, were used in the second phase of this experimentation. The use of these types of materials was with the intent to investigate the oxidation potential of these common materials when used as cover material for arid countries.

Column reactors were prepared and designed to test a material having a general set of soil characteristics, atypical from the artificial materials that had been used by several other researchers and those reported in the literature, aimed at testing landfill oxidation. To fulfil the objectives outlined in Section 1.3, a system of column reactors was designed and then built by the workshop. However, to use these reactors in a laboratory requires addressing safety concerns. Testing a mixture of highly combustible and flammable methane and oxygen gases in the university laboratories, particularly when unattended, assumes more important consideration than and above all of the entire testing requirements. Therefore, considerable time and effort were taken to satisfy the university health and safety requirements. A risk assessment report to

satisfy these requirements was prepared and presented to the safety department of the university, which then had been studied and approved (Appendix D). To fulfil these requirements, some limitations had to be accounted for. It was deemed necessary to have low flows in the tubes and the reactors in case a full leakage would mount up during a period of a weekend. The same holds true with small size reactors so that a methane build-up inside them would not occur. These challenges meant expensive flow meters to be used in order to accommodate the low flows. It is also worth noting that the low flow makes it less noticeable and harder to find leaks; while small tubes meant that they could easily be blocked.

A system of reactors was set up at Cassie Building's pilot laboratory on the ground floor, in which six columns were connected together, as shown in Figure 3.2. Each set consisted of two identical column reactors (as duplicates) of 3-cm diameter, and 13 cm in height, connected separately, filled with identical materials, and fed with identical input variables to ensure repeatability and better accuracy of the experiment. Each of the sets (three sets of two reactors system) was filled with up to 9 cm of different soil/sand ratios, 9:1, 1:1, and 1:9, placed over a steel mesh with the columns sealed from top and bottom with rubber caps. In these sets, clean sand of 2 mm was added to the soil material composed of 16, 26, and 57% clay, silt, and sand of granular makeup, respectively. Each cap had open orifices for the purpose of introducing and measuring the input and output variables. Specific low flow rate meters were placed for the inputs of methane and oxygen to the system. The flow meter that was used was from Bronkhorst (F-201CV-020-AAD-22-V Digital Mass Flow Controller, one for methane and one for air) with the BRIGHT B2 IP40 9p SubD display and kit. In addition, it was necessary to avoid any tubes or materials that would absorb methane in the process, thus Tygon tubes (B 44-EU) were used of different lengths and diameters depending on the need with connectors. The air and methane supply came from cylinders in the laboratory provided by BOC, fitted with specific regulators for gas and methane (Gas Safety UK (B J Industries Ltd) PART No. 99-005 0-4 barG and PART No. 99-011S 0-4 barG, respectively) with push-fit connectors and an EasiDaptor. On the other hand, the air and methane gases were distributed using a Manifold (made of Brass Screwed-Bonnet Needle Valve, 1/4 in. Swagelok Tube Fitting, Panel Mounting, Cat. B-4JN-PM, Brass

Swagelok Tube Fitting, Union Elbow, 1/4 in. Tube OD, Cat. B-400-9, Brass Swagelok Tube Fitting, Union Tee, 1/4 in. Tube OD, Cat. B-400-3, and Brass Swagelok Tube Fitting, Union Cross, 1/4 in. Tube OD, Cat. B-400-4).

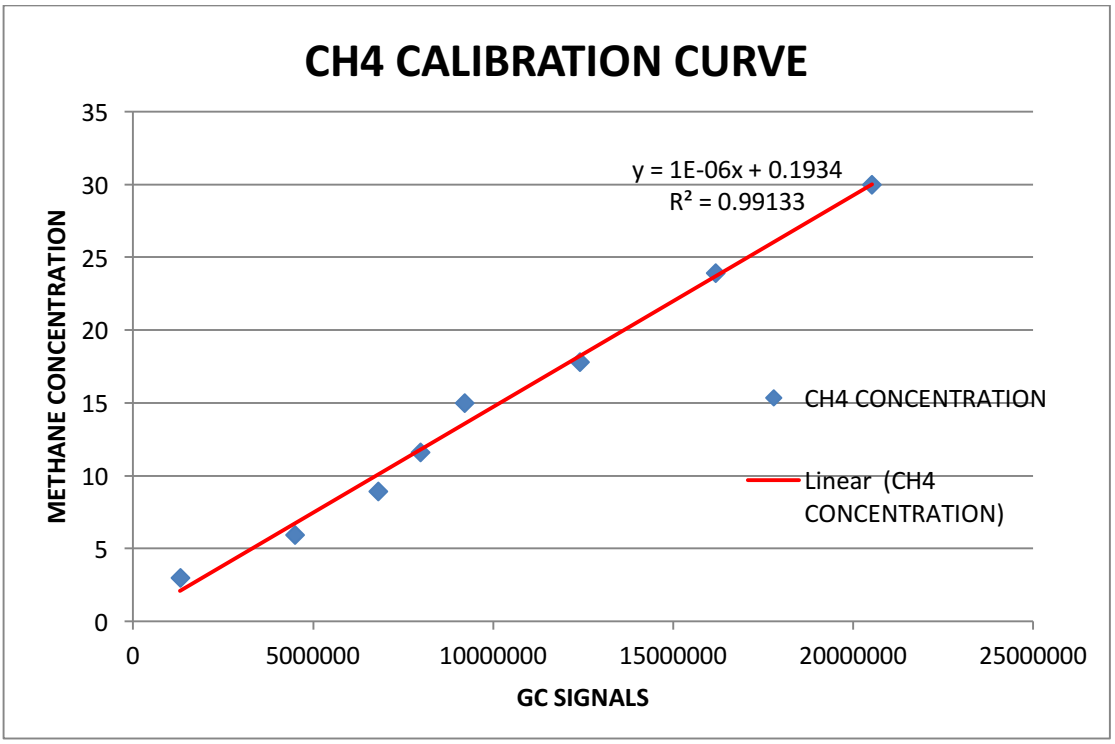


Figure 3.1: Typical calibration accuracy curve for the GC analyser.

The column reactor system was used as a continuous flow setup that can simulate landfill variables more closely than the batch tests, and can allow the flexibility of altering the input variables at any time. For the repeatability requirement, it would have been more accurate to build three identical sets, or even more, but to have done so, would entail higher costs for the investigation. Thus, after a long process of looking into the university safety and considerable checking of the system for leaks and equipment for accuracy, the system was started at room temperature of 22+/- 2^{o} C. Methane gas and standard air, in a ratio of 35% to 65%, respectively, were introduced through two separate flow meters into a bottle containing water to get full mixing and moisturising of the gases at the same time. The mix was then fed, passing through bubble counters to each individual reactor at a rate of 2.5 ml/min and 0.8333 ml/min for air and methane gases, respectively. Outputs of the reactors were taken from the

top of the reactors, starting after one day of running, using other bubble counters with infrared electronic counter. GC analyser with FID was used in the experiment for measurements. The variables were observed, monitored, and recorded. Since this system was operated continuously and fed with continuous gases, the reactions developed quickly and continuously; therefore, measurements were taken every 12h to one-day interval.

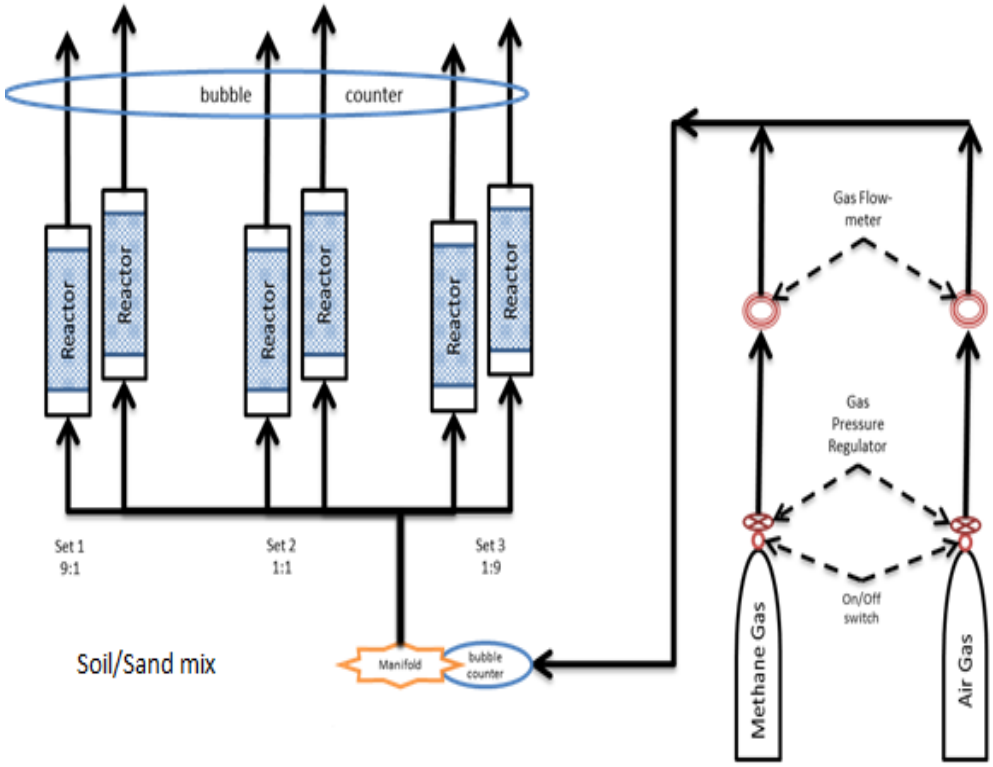


Figure 3.2: Schematic diagram of continuous flow reactors filled with different sand/soil ratios.

3.4.2 *Total organic matter (OM), total organic carbon (OC), total nitrogen (TN), moisture content (MC), and pH level*

At the outset of the experiment, the acidity/alkalinity of the soil and sand mixtures were measured in accordance with the ISO standard 10390:2005, by taking approximately 10 ml of the soil mix, immersed in a beaker filled with 50 ml of distilled water. The mix was then stirred for few minutes, left to settle for 10-15 min, then pH meter probe was immersed in the liquid for measurements. Each time the pH meter

was used, a calibration procedure was taken, using standard buffer solutions, set at pH 4 and 7.

Moisture content (MC) is an important parameter in the system of methane mitigation, particularly for nutrients transport and for carrying bacterial refuse. Therefore, to measure MC, ASTM standard (D4442) was used, where a 30-g sample was taken for each one set of the sample mixtures, measuring first the weight of an oven Petri dish, and then the weight of the sample mix with the dish. The sample and the Petri dish were then placed in a furnace set at 105°C, weighed again after one hour in the oven, to find the weight of the lost moisture due to the oven evaporation. The same procedure was performed on the mixtures of every set for each sample.

Similar procedure was taken to find the weight of the organic matter (OM) existing within the sample mixtures. Ten grams of sample mixtures was taken from each one of the three sets, oven dried to relieve the samples from moisture. This was achieved by placing the samples in a known weight of a Petri dish, placed in an oven set at 105°C, then the weight of the dried samples was calculated. The samples in the Petri dish were again placed in the furnace, set this time at 550°C, for a duration of ten minutes to burn and evaporate the organic contents in the samples, then, the weight of the samples was measured, and the OM was calculated. Total organic carbon was calculated using the procedure adopted by Jimenez and Garcia (1992), in which total organic matter was correlated with total organic carbon (modified Van Bemmelen factor, Pribyl, et al., 2010). These calculated values were also compared with laboratory procedure, where a combustion method using a Carlo Erba CN analyser (Flash1112 series) was used. Samples were ball milled for homogenisation at milligram level. Around 30 mg of milled soil was weighed in tin capsules using a 6- decimal balance and then pressed before being analysed simultaneously for total carbon and total nitrogen.

3.4.3 *Porosity and soil characterisation*

The porosities of the samples were measured according to equation 2.3 of Section E.5 (Appendix E). Thirty grams of soil/sand mixtures was measured from each sample of each set of the experiment, placed in a known weight of Petri dish, and then oven dried. The weights and volumes of the dried samples were measured; then each sample was immersed in distilled water of known volume, contained in graduated cylinder, to measure the volumes of the solid mass of the sample. By using equation 2.3, the porosity of the samples was then calculated. Soil particle size distribution and characterisation for soils samples used in the batch experiments were performed using wet sieving method in accordance with British Standard (BS-1372-2-1990), utilizing the mechanical sieving equipment at the Geotechnical Engineering and Material Laboratory at Drummond Building. For the soils used in column test, Laser Diffraction Particle Sizer apparatus was used, where thirty grams of soil samples were suspended in water and passed through the flow cell of a Laser Diffraction Particle Size Analyser (Beckman Coulter LS13320). In this process, the intensity of light scattered particles was found to be directly proportional to the sizes of the particles. For this analysis, which required special equipment and needed better accuracy, the analysis of the soil and the total nitrogen in the samples were entrusted to an outside laboratory (The Forest Research Centre for Ecosystems, Surrey, UK).

3.4.4 *Column test for oxygen penetration*

Dust fallout in arid climates limits air from diffusing into the top cover layer of landfills, preventing oxygen, the very vital element in methane mitigation, from reaching the assimilating bacteria in the soil. In addition, regulations have mandated that only low permeability top cover layers could be used on landfills to prevent rainwater from forming leachates in the sub layers of the landfills. These facts limit the use of any standard passive or new capture methods for methane control. Therefore, if a new system of delivery can be devised to make oxygen available to reach into the deeper layers of the landfills, without disturbing the blocked or regulated top cover layer of the

landfills, then that would create a barrier system beneath that top cover layer, hence, mitigating methane more efficiently.

In order to investigate the effects of deeper presence of air inside the layers of the landfills on the performance of the methane-assimilating bacteria, instead of relying on the atmospheric supply of oxygen, another continuous flow reactors experiment was devised. Although it is well-known that deeper oxygen supply would provide more oxygen for the bacterial needs inside the layers of the oxidation layers; albeit, the efficiency of this delivery, that could warrant investment into new mitigation design, is in need of further investigation. This experiment, was therefore aimed at investigating the oxidation performance of the methanotrophic bacteria under the supply of air at different levels inside the soil layers.

In this experiment, three sets of 60-mm diameters, and 75-cm high reactors were constructed by the workshop and designed to include openings on each side of each reactor; however, each opening was installed to be used at different level on these sides (Figure 3.4). The locations of orifices were constructed at 0, -13 cm, and -26 cm, beginning from the top level of the soil mixture in the columns, moving downwards; while other openings were also made at the bottom and top sealing caps of the columns. The side openings were meant to supply air; while the bottom and top sealing cap openings were meant for the supply of methane and collection of output gases, respectively. A duplicate for each set of these columns was constructed and the sets were then placed in the Cassie Building's pilot laboratory at the ground floor. The schematic diagram and the setup of this experiment are shown in Figures 3.3 and 3.5, respectively.

Each of the columns was filled with identical material of ground soil, mixed with compost material in a ratio of 1:1 with a wet weight of 747.7 g, and the moisture content of 29.0–31.0%, and placed on top of a steel mesh plate inside each column. All columns were kept at the same room temperature (22+/-2°C), with all other surrounding conditions being monitored and maintained as the same. Methane gas was introduced from the bottom of each column, simulating an upward surge of

landfill's methane movement; while oxygen was introduced at different levels of each set of the column tests, simulating different levels of penetration of oxygen into a landfill. A continuous feed of 1 ml/min methane was introduced at the bottom of each set of the columns, and a 3.333 ml of air was fed into the side orifices of the columns at levels 0, -13, and -26 cm down from the top surface of the soil mix, as sets 1, 2, and 3, respectively. Moreover, input and output gas flows were measured using bubble counters, flow meters, and analysed with a GC-FID. The bubble counters were bottles filled with distilled water, allowing gases to pass through, and in their way out, they made continuous bubbles that could be counted using electronic sensing counters.

The air and methane supply came from cylinders in the laboratory provided by BOC, fitted with specific regulators for gas and methane (Gas Safety UK (B J Industries Ltd) PART No. 99-005 0-4 barG and PART No. 99-011S 0-4 barG, respectively) with push-fit connectors and an EasiDaptor. While the air and methane gases were distributed using a manifold (made of Brass Screwed-Bonnet Needle Valve, 1/4 in. Swagelok Tube Fitting, Panel Mounting, Cat. B-4JN-PM, Brass Swagelok Tube Fitting, Union Elbow, 1/4 in. Tube OD, Cat. B-400-9, Brass Swagelok Tube Fitting, Union Tee, 1/4 in. Tube OD, Cat. B-400-3, and Brass Swagelok Tube Fitting, Union Cross, 1/4 in. Tube OD, Cat. B-400-4).

Gases measurements were taken daily similar to the procedure followed in section 3.4.1, and the measurements of porosity, OM, MC, TN, pH and soil characterization were all taken according to the procedures outlined in sections 3.4.2 and 3.4.3.

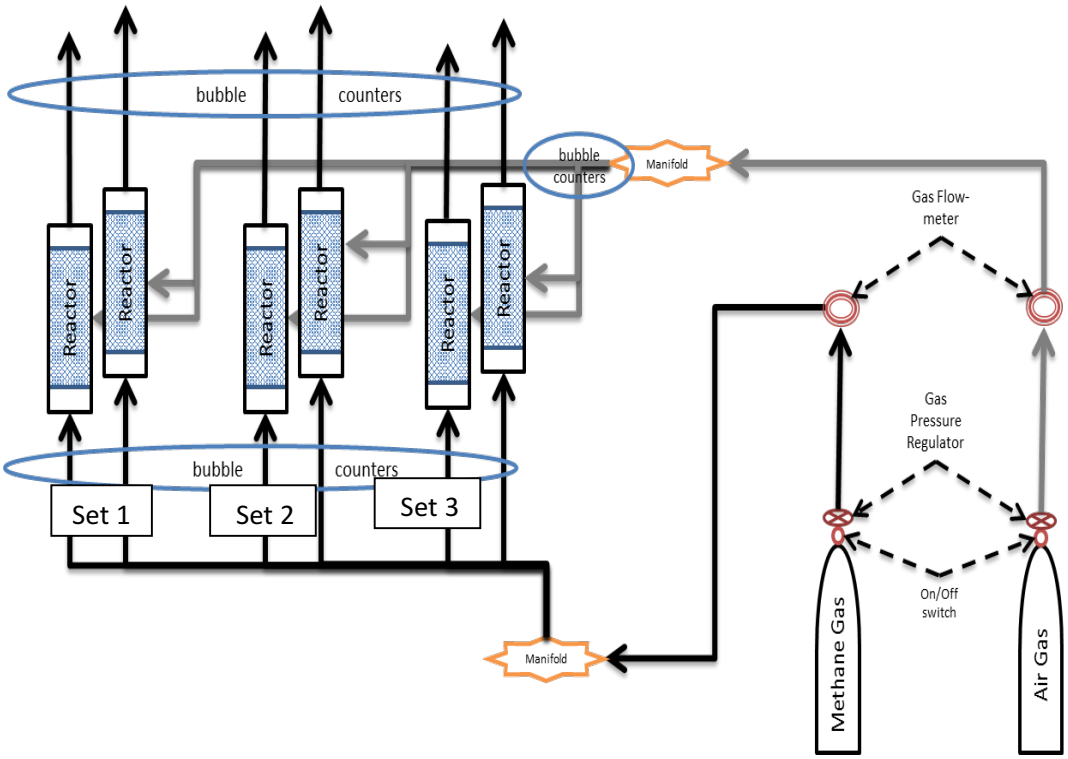


Figure 3.3: Schematic diagram of the column experiment, testing for the effect of oxygen penetration on the methane oxidation.

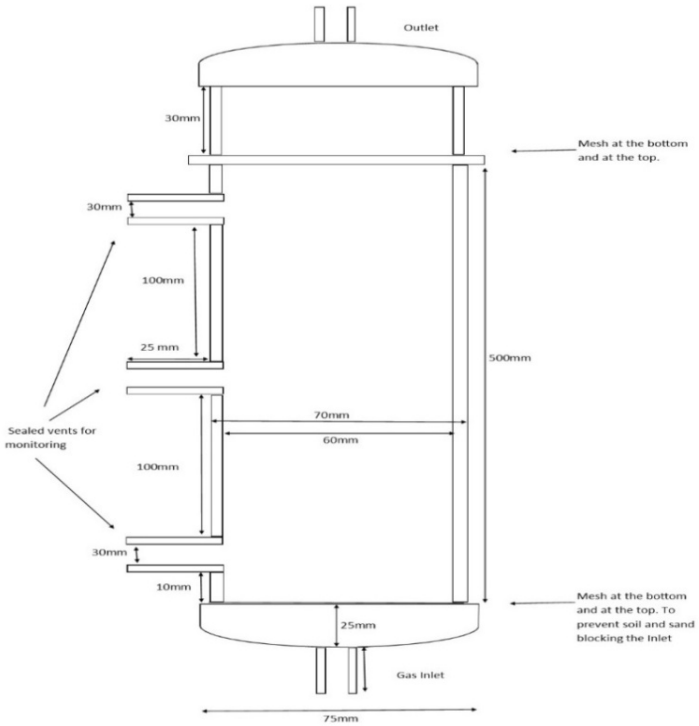


Figure 3.4: Column reactor design.

Figure 3.5: The column experimental setup at Cassie Building's pilot plant, University of Newcastle.

Chapter IV

Experimental Investigations: Results and Discussions

4.1 Batch tests results and indications

The batch experimental results are shown and discussed in the following subsequent sections.

4.1.1 *Methanotrophic activity in different types of soils*

Data collected from the batch reactor chambers showed little or no oxidation reactions by the methanotrophic bacteria present in the bulk of the landfill samples collected from several areas described in Table 3.3. From these data, it was noticed that the samples showed little change in methane presence in the reactors with the passing of time, except for the leachate samples, which showed an increase in methane production due to their original high landfill methane exposure and toxicity, producing the results shown in Figure 4.1. The results in this figure indicated different measured methane consumption at the outset of the experiment, due to bacterial activities occurring after one day of incubation period before the start of the measurements. Two other samples, Land-GCP-1 and Land-LCP in Table 3.4, taken from different locations of the local landfill soil, showed little oxidation reaction; however, all the other samples did not react to the presence of oxygen due to the nature and granular makeup of the samples. When the same samples were made smaller and then diluted by nutrient solution (Table 3.5), this change allowed a higher ratio of oxygen to sample surface area to exist, allowing higher oxygen penetration to the inner particles of the samples to materialise, hence, giving rise to a better methanotrophic bacterial reaction to the presence of oxygen. This resulted in the depletion of oxygen present in the reactors, where the other methanogenic bacteria started the anaerobic production of methane, as indicated in Figure 4.2.

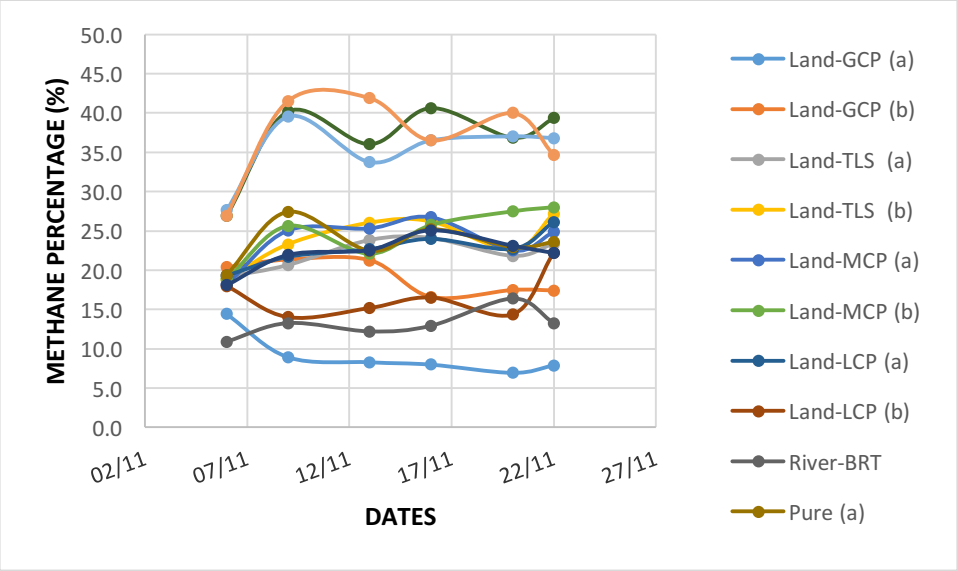


Figure 4.1: Methane percentage consumed by batch soil samples (in duplicates, a, b) as defined by Table 3.3, (year 2012)

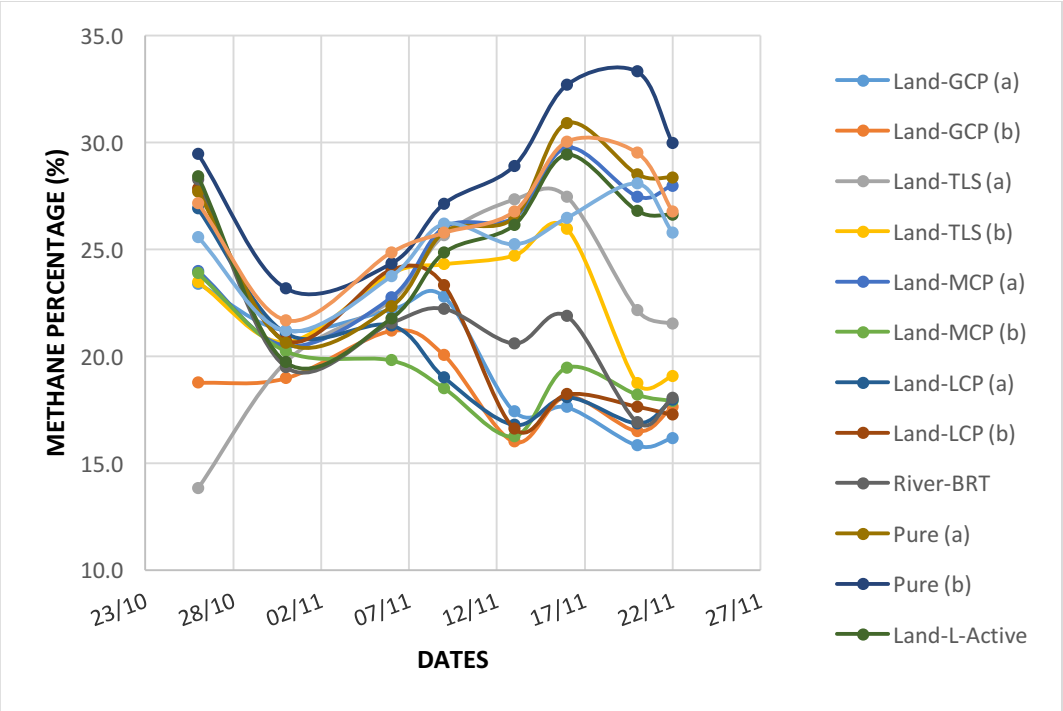


Figure 4.2: Methane percentage consumed by batch soil samples (in duplicates a, b) with increased media solution as defined in Table 3.5, (year 2012).

Increasing the ratio of oxygen penetration and diffusion in relation to the volumes of the samples, with appropriate nutrients present by amounts prescribed in Table 3.5, resulted in mixed outcomes (as indicated in Figure 4.2). Methane consumption rate increased by approximately 30% over a 10-d time interval for most of the tested samples, which then began to decrease continuously for all of the samples, except for samples Land-GCP, Land-MCP, River-BRT, Land-L-Old, and Land-Mix. These results can be explained by the different makeup, such as in grain size, orientations, volume of voids, presence of inhibiting substances, etc. (Sections E.4, E.5, E.23, Appendix E), in the microstructure of the soil samples, consequently, resulting in each, having the distinctive ability to hold different levels of oxygen. Although the samples were diluted with water and other soluble materials, the addition of nutrient solution is particularly important, because it loosened the bonds of the medium, allowing for better molar diffusion of the nutrients to supply the bacteria with nutrients (Section E.13, Appendix E). As for the earlier samples listed in Table 3.4, while they were not diluted with more media solution, they were not able to hold oxygen, and hence, allowed for poor oxidation performance. All confirm the well-researched fact, that soil types in landfill cover must possess high porosity values in order for them to utilise the full potential of the oxidation of the methanotrophic bacteria.

4.1.2 *Bacterial behaviour in differently exposed soils to CH_4*

When designing landfill bio-covers, little was researched to identify the time period that affects methanotrophic bacteria to be active after the immediate installation of the cover. Due to the high amount of methane emitted into the atmosphere, estimated to reach 500–800 metric tons of CO_2 equivalent per year from landfills alone (Intergovernmental Panel on Climate Change, IPCC, 2007), and given the ever increasing human and land filling activities, therefore selecting a type of soil cover for bio-covering or bio-filtration that will immediately enhance methane consumption is essential. Accordingly, low and highly exposed methane soils were compared in order to understand the time effect of these biologically active and non-active soils, simulating active and non-active covers on methane elimination. These tested samples are described in Table 3.6, and the results of the observations are plotted in Figure 4.3,

for soils obtained from active landfill and the non-active garden grounds (university campus ground soil).

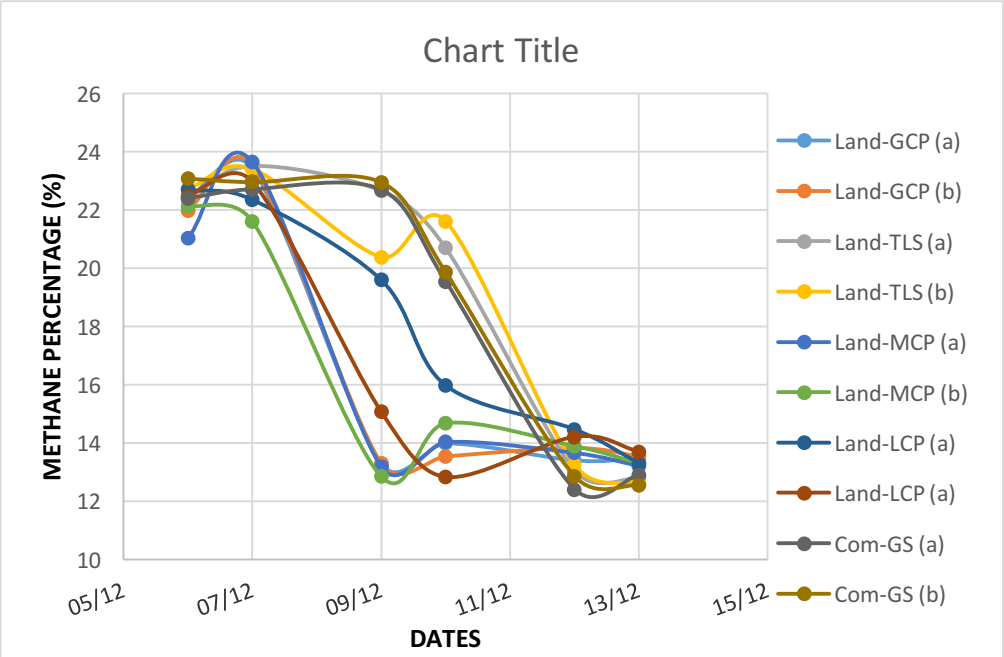


Figure 4.3: Comparison of methane percentages consumed by batch experiments of samples (in duplicates a, b) as defined in Table 3.6 (year 2012).

It was observed that a time lag between active and non-active soils, when used in the batch incubators, was remarkably clear, as it took approximately 4 d for the methanotrophic bacteria to become active in consuming methane in the non-active soil medium. The samples from the lower side location of the landfill, a distance away from the methane collection pipe as well as the clay and sandy soil samples from the Newcastle University grounds were both seen to have reduced methane from approximately 22% v/v to approximately 13% v/v. Both samples exhibited a time lag of 4 d for the bacteria to become fully active. The other samples, namely the soil sample taken near and on top of the methane collection pipe of the landfill, consumed the same amount of methane. Conversely, they exhibited an immediate response of consumption; while the samples collected near the leachate pipe had an intermediate response. This latter intermediate behaviour could be due to the unfavourable and toxic habitat to the methanotrophic bacteria in and near the leachate section of the landfill to multiply normally.

It was also noticed from the shape of the curve of samples Com-GS-a and Com-GS-b of Figure 4.3, that once the bacteria had been activated, the rate of consumption of approximately 7.15 v/v of methane per day was the same for both the low and the highly exposed samples. This would imply that the bacteria type and the quantity of the two differently exposed samples had reached the same rate of activities once allowed sufficient time, with only a time delay at the beginning.

Typical landfill sites are filled daily with municipal wastes, deposited from collection trucks into landfill cells, with each cell, being sealed at the end of the day with earth soil covers Figure 4.4. These earth soils' covers of 6- to 12-in. thickness are placed either one on top of the other or side-by-side, for purposes of preventing odour and health hazard from contaminating the surrounding environment. However, these covers were not exposed to methane at the time of this operation, which only means that some time must pass before the methanotrophic bacteria could start assimilating methane. In a year time, in approximately 300 working days, they would place 300 new daily covers, with unexposed earth's soils, which could allow unassimilated methane to escape to the atmosphere. Each cover would permit a time of 3–4 d of unchecked methane, emerging from old and new cells, alike, to migrate from cells to this daily cover, then, to the atmosphere (Figure 4.3). Given the fact that global estimation of landfill gas production is an imperfect science, usually based on the amount of waste available for the biodegradation of biological component of the waste, using theoretical models (UNEP, 2010), an estimate of methane escaping due to this lag of methanotrophic activity could be calculated. Taking the global estimated methane production to be of 760 MMTCO2-eq per year (Table 1.1) and the approximate 300 unamended landfill covers placed during the working days, subsequently, this could translate into 2.53 MMTCO2-eq of methane release per each earth cover per year, globally. Consequently, 3–4 d of unassimilated methane would produce a total of 7.6 to 10.1 $MMTCO_2$-eq a year, which would escape unoxidised into the atmosphere. For landfills to operate for approximately 20 years before final closure, this produced amount of methane per day globally would be considered an important factor for landfill managers when covering cells on the daily basis with unexposed covers, according to the US-EPA landfill model (Figure 4.5). Figure 4.5 shows an indicative

graph generated by the US EPA LandGem model to estimate methane generation throughout the life cycle of a landfill. The figure shows that landfill gas production increases continuously and incrementally up to closure time, after which landfill gas production would decrease rapidly. Nevertheless, if control methods were to be installed for LFG recovery, the curve would take a different shape. This time lag, if not addressed by placing a pre-exposed cover, could result in the escape of methane into the atmosphere, and consequently, could translate into either carbon international taxation costs, or cover redesign costs to readjust landfill cover layers.

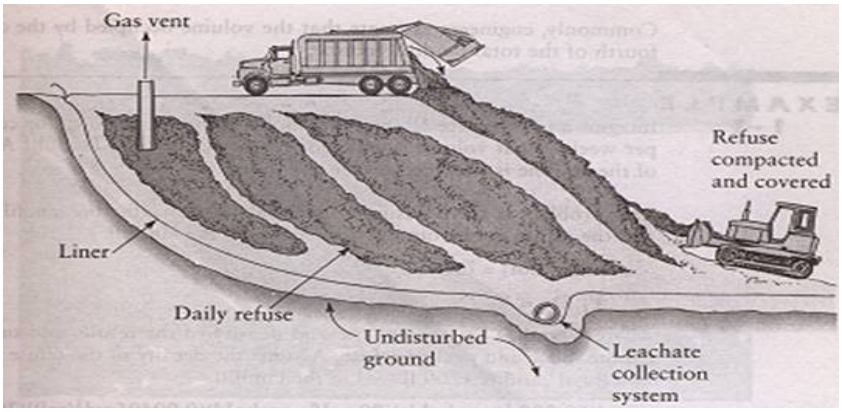


Figure 4.4: Schematic diagram of typical landfill cells and coverings (source: Mishra, The Constructor Engineering Org.).

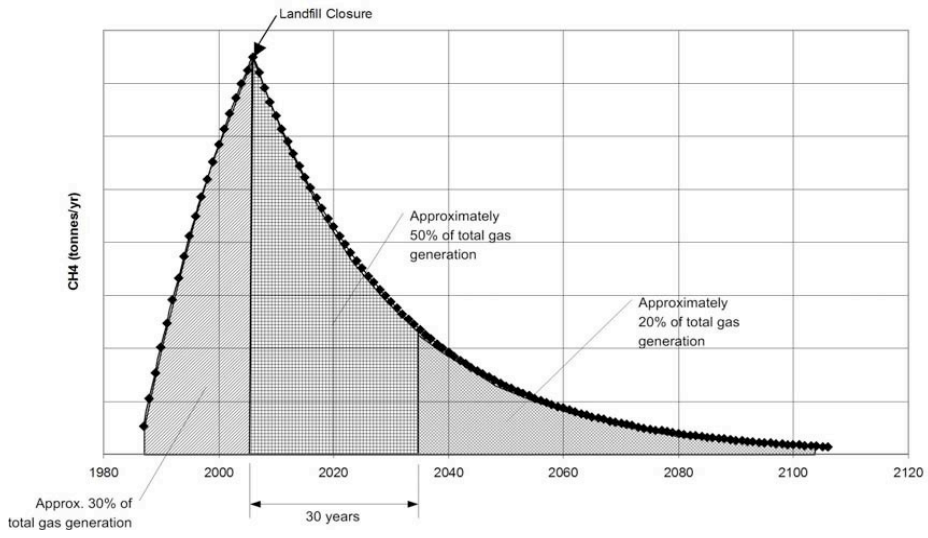


Figure 4.5: Indicative methane generation curve in landfills before and after closure (US EPA LandGEM, 2005).

4.1.3 *Bacterial response to oxygen availability*

Another mechanism introduced to these series of batch tests is the shaking of the samples. This process allowed physical bonds of the samples to be loosened, creating micro voids in the samples, therefore allowing better air penetration into the soils and faster molar diffusion, subsequently, resulting in uniform methane consumption in the presence of nutrients. In this instance, oxygen was delivered by forcing air to penetrate due to mixing of the contents of the reactor and due to molar diffusion, despite the condition of the samples. This process allowed the bacteria to consume more methane and to continue for a longer time span, until reaching a constant methane percentage average.

As shown in Figures 4.6 to 4.8, the shaking action performed on the samples, regardless of their soil types, had a profound effect on the consumption of methane and in essence had allowed the oxygen to penetrate through the soil particles and distribute the oxygen moles directly to the methanotrophic bacteria, and allowed methane to dissolve into the water, reaching the residing bacteria in the soil. However, this shaking mechanism did not provide much help after the oxygen was, and after anaerobic action took effect, at which methane concentration started to rise again for almost all the samples, even more so, under the shaking condition, and less when under static state (Figures 4.6 and 4.7). To offset this behaviour, a dose of oxygen was injected into the reactors for all the samples and for the shaking and static samples at time intervals, indicated in Figures 4.6, 4.7, and 4.8. The oxygen dose lasted for approximately five days for both conditions before methane concentration started to rise again. Another dose was injected, which further helped in the reduction of methane for both conditions.

This dramatic reduction of methane when oxygen was introduced is evidently an important observation, regardless of the other parameters present in the samples. Moreover, the process of continuously dosing oxygen into the incubators in a sustained measure produced an approximately continuous linear relationship in the reduction of methane with time, as evident from Figures 4.6-4.8. Also, it is worth mentioning that

the linear relationship has the same rate of methane consumption, estimated at 0.90% v/v per day for both, before and after the introduction of oxygen, in case of the static conditions as shown in Figure 4.7. However, the rate of consumption was much higher for samples under shaking, estimated to equal 2.0% v/v per day before oxygen was depleted for the samples under static conditions (Figure 4.7), a rate of which was double for the shaking condition over the non-shaking, for the same stretch of time.

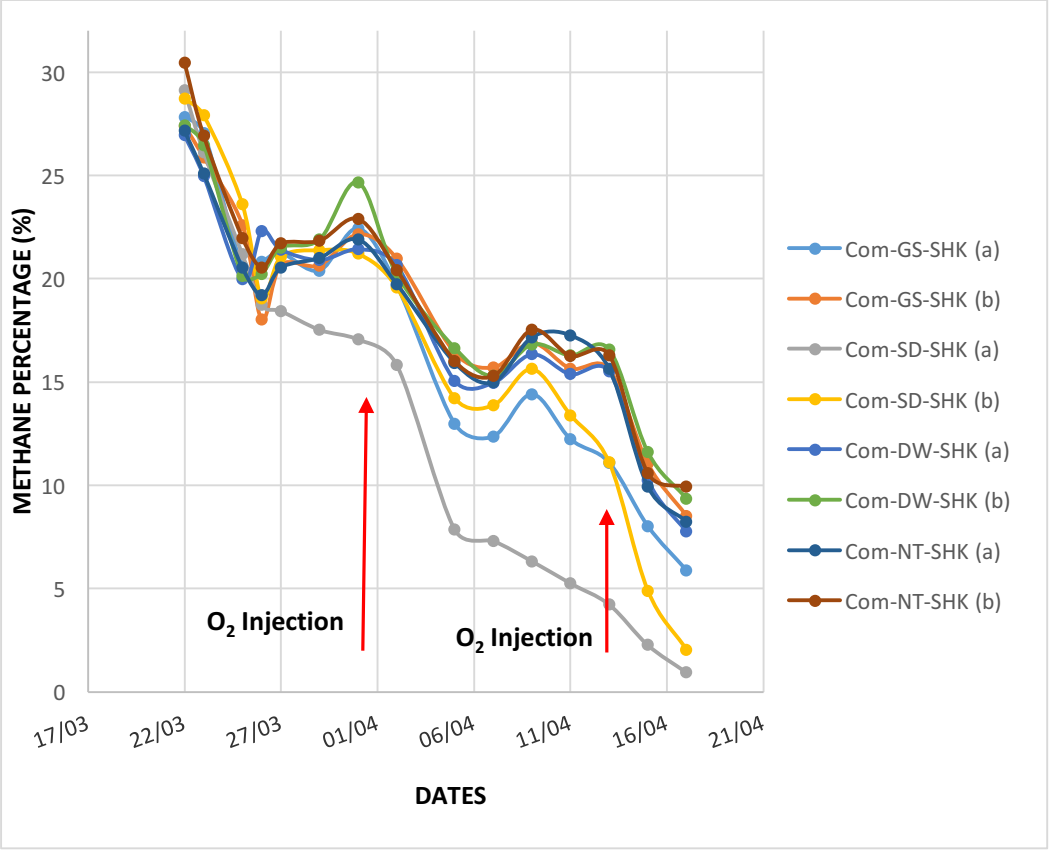


Figure 4.6: Methane percentage consumed by batch soil samples (in duplicates a, b), under shaking condition and after introduction of oxygen (Table 3.7, oxygen injected on 31/03, and 11/04/2013 at the arrows).

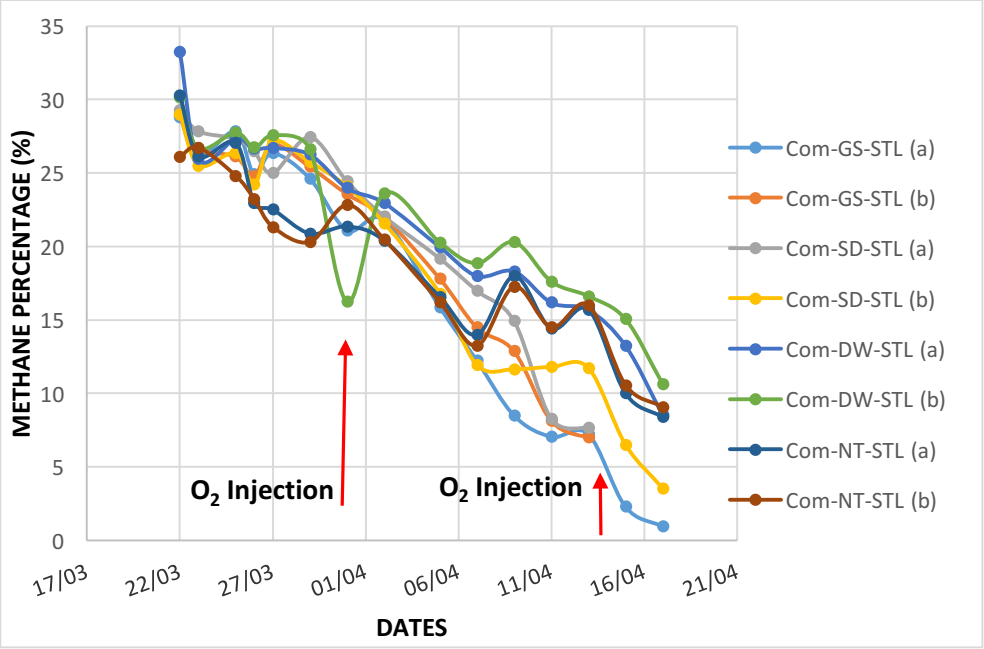


Figure 4.7: Methane percentage consumed by batch soil samples (in duplicates a, b), under static conditions and after introduction of oxygen (Table 3.7, oxygen injected on 31/03 and 13/04/2013, at the arrows).

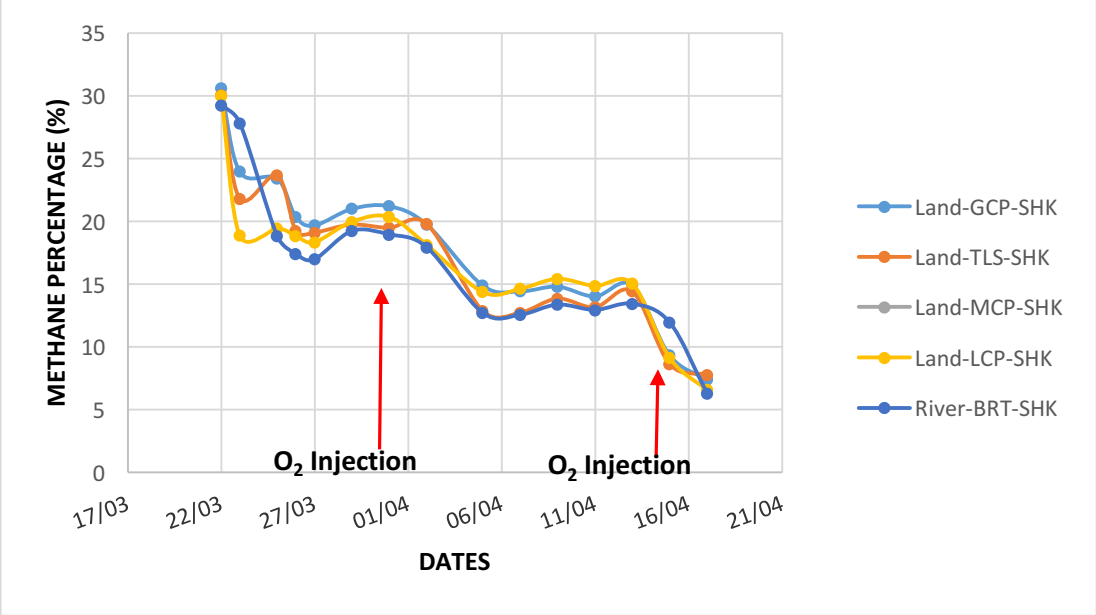


Figure 4.8: Methane percentage consumed by batch landfill soil samples (in duplicates a, b), under shaking conditions, with added media solution and after an introduction of oxygen (Table 3.7, oxygen introduced on 31/03 and 13/04/2013 at the arrows).

Reactors with only room air and methane, as control reactors, having the same v/v ratios as in each of the test sets, showed approximately a drop of 1% of the methane volume at the start of measurement, and no appreciable change in the relative volumes afterward, in which this drop was accounted for in the results of these batch tests. This small drop could be attributed to molar interactions, or to the possible interaction with trace gas elements present when room air was introduced into the reactors, at the beginning of the experiments, since methane and oxygen are reactive gases with other gases and with the possible trace elements present in the reactors.

For the sake of comparison, rates of maximum oxidation of the samples described in Table 3.6, and shown in Figure 4.3, were calculated and presented in Table 4.1, along with some results of maximum oxidation rates obtained for published literature experimental results. Table 4.1 presents the average oxidation rates of each set of the duplicate reactors of the batch experiment conducted in this research, indicating that exposed landfill soils to methane had higher rates of oxidation and revealed oxidation rates of 117.2 and 113.0 μg CH_4 g^{-1} h^{-1} for Land-GCP and Land-MCP, respectively, higher than the common ground soils that had little methane exposure. These common ground soils were Com-GS, averaged at a rate of 69.3- μg CH_4 g^{-1} h^{-1} in all samples having initial methane concentrations as indicted in the table. In a comparison study for this batch soil samples, Schuetz and Kjeldsen (2004) showed that the testing of loamy sand soil produced a maximum oxidation rate of 118- μg CH_4 g^{-1} h^{-1}, when subjected to 15% (v/v) initial methane concentration. This soil has similar soil composition to the soil investigated in the batch experiment shown in Figure 4.3. Also, another published batch test result, using humic soil till showed a maximum rate of oxidation of 86.4- μg CH_4 g^{-1} h^{-1}, when methane concentration was introduced initially at 10% (v/v) (Figueroa, 1993). These published results are of the same order of magnitude as the results of the batch experiments conducted in this research. Nonetheless, results should be taken to be symptomatic, considering the difference in the granular makeup of the soils, relative to operating temperature, chemical composition, etc., of each set of experiments.

Sample/Soil	Initial Methane Concentration (average) %(v/v)	Maximum Oxidation (average) μg CH_4 g^{-1} h^{-1}	Temperature C^o	References
Land-GCP	22.16	117.2	22 +/-2	
Land-TLS	22.57	64.7	22 +/-2	
Land-MCP	21.57	113.0	22 +/-2	
Land-LCP	22.59	74.4	22 +/-2	
Com-GS	22.73	69.3	22 +/-2	
Loamy sand	15.0	118.0	30	Scheutz and Kjeldsen, 2004
Humic soil till[a]	10.0	86.4	30	Figueroa, 1993

[a] A fine textured compost, made with yard trimmings using a soil conditioner.

Table 4.1: The batch tests in comparison with published literature results

Other samples with various mixing and additive combinations and under the condition of shaking and still conditions on a bench were also investigated. The results are shown in Appendix B, which exhibited the same experimental behaviour as that of the data plotted in Figures 4.6 and 4.7. In addition, some samples were also subjected to inhibiting substances to be able to show the effect of inhibiting substances on methanogenic bacteria, as well as on methane elimination.

4.2 Column tests of soil in arid environmental climates

Tests of sand and soil materials used in column experiments were intended to simulate landfill cover in desert environment, similar to that existing in Kuwait. The sand soil materials present in Kuwait are abundant and have compositions indicated in Tables 3.1 and 3.2, showing high rates of medium and fine sand particles, which produced an average hydraulic conductivity of 4.0 X 10^{-7} m s^{-1}. The cover soils of the country were classified as sand and sandy loam soils.

The particular characteristics of the soil/sand mix of the materials used in this experiment are shown in Table 4.2, where the soils encompassed the range of mixes present in Kuwait soils, ranging from 0- to 2-µm clay, 2- to 63-µm silt, and 63-µm to 2.0-mm sand, a mix of fine to coarse sizes. The pH values of all the materials in the mixes of the sets, were all slightly alkaline at 7.1, 7.29, 7.28 for Sets 1, 2, and 3, respectively, with an average pH of 7.19, which is of a close level to the average pH values of Kuwait soil at 7.45, shown in Table 3.1. OMs were variable in the test set materials, ranging from 18.56% (wt/wt) for the high ratio of soil to sand (9:1); while it was merely 0.67% (wt/wt) for the low soil to sand ratio (1:9). These low OM rates are expected, since ground soils contain more OM than the clean added sands of 2.0 mm in diameter. On the other hand, moisture contents (MC) in the materials of the sets showed a value of 26.08% (wt/wt) for the high soil ratio and 2.45% (wt/wt) for the low soil ratio. Again predictable, because of the higher moisture retention characteristic of fine-grained soils vs the low moisture retention characteristics of the sands of larger diameter, which allowed high amount of moisture to be present in the set of 9:1 soil to sand (sandy loam), remaining drier for the low ratio of soil to sand (1:9) (sand). The average hydraulic conductivities of the soil samples in the sets (Structx, 2014-2015), shown in Table 4.3, indicate that they are little higher than the average hydraulic conductivity of the Kuwaiti soil.

Table 4.2 presents a comparison between the soil and sand mixes used in the sets of this experiment, with results obtained from published literature of clay soil showing relative close relationship. The pH value used obtained by He et al. (2008) was 7.11, a slightly alkaline clay soil, which was in close comparison with the soil and sand materials used in these sets of the experiment. It also exhibited a granular soil makeup of typical clay soil at a range of < 0.02 mm, a little finer in granular makeup compared to the range of granular makeup used in the experimental sets of this study.

The test results on these materials when methane gas and standard air mix were introduced into each experimental column from the bottom, at a ratio of 35 to 65% with a flow rate of 2.5ml/min, respectively, are shown in Figures 4.9, 4.10, and 4.11. In these figures the percentage of the methane oxidation consumption with respect to

methane input was calculated for Sets 1, 2, and 3, respectively. Table 4.3 shows also these comparative results.

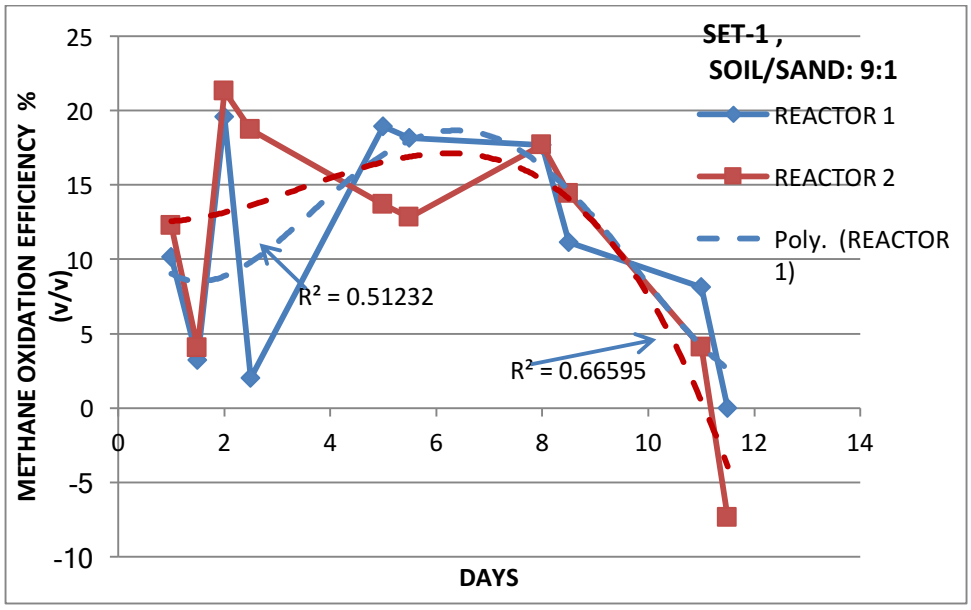


Figure 4.9: Methane oxidation efficiencies for column reactors set 1.

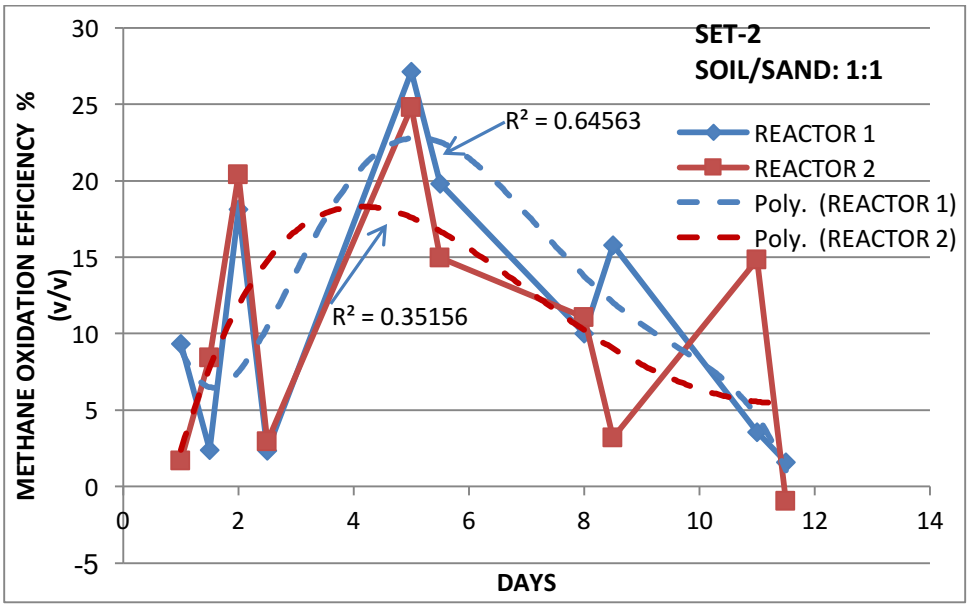


Figure 4.10: Methane oxidation efficiencies for column reactors set 2.

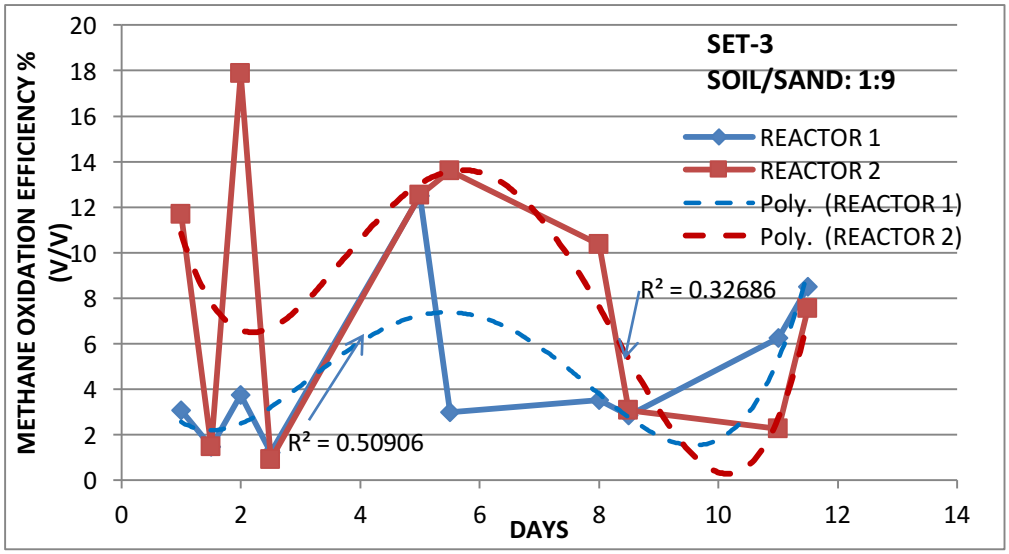


Figure 4.11: Methane oxidation efficiencies for column reactors set 3.

Exp.	Type of Mix	Soil granular characteristics	Porosity (%)	pH	Organic Matter (OM) (%) (wt/wt)	Moisture Content (MC) (%) (wt/wt)	Total Nitrogen (TN) (%) (wt)/(wt)	Total Organic Carbon (%) (wt/wt)	C/N Ratio
Set 1	Soil/Sand 9:1	Clay: 13.0% Silt: 20.0% Sand: 67% (Sandy Loam)	17.5	7.10	18.56	26.08	0.763	11.388	14.93
Set 2	Soil/Sand 1:1	Clay: 9.0% Silt: 12.0% Sand: 78.5% (Loamy Sand)	25.0	7.29	3.62	10.13	0.124	1.859	14.99
Set 3	Soil/Sand 1:9	Clay: 1.8 % Silt :2.5 % Sand: 95.7% (Sand)	43.7	7.28	0.67	1.45	0.024	0.403	16.79
He et al., (2008)	Clay soil	2–4 mm 9.3% 0.02–2 mm 27.1% 0.002–0.2 mm 38.7% < .002 mm 24.7% (Clay)	----	7.11	0.54	14.0	0.0051	0.06	11.76

Table 4.2: Soil and sand mix characteristics, simulating desert materials (values are average of two reactors).

Experiment	Type of Mix	Average Hydraulic conductivity (m/s)	Methane Loadings kg CH_4 m^{-2} day^{-1}	Maximum Oxidation %v/v	Temp. (°C)
Set 1, (9:1)	Sandy loam	7.19 x 10^{-5}	1.075	11.1	22+/-2
Set 2, (1:1)	Loamy sand	1.56 x10^{-4}		10.6	
Set 3, (1:9)	Sand	1.76 x10^{-4}		6.4	
He et al. (2008) Exp.	Clay Soil	--	0.260	10	30+/-1

Table 4.3: Maximum oxidation in comparison with literature findings

The results shown in the figures and in Table 4.3 were obtained as a consequence of an interaction between a continuous feed of mixed air (21% oxygen) and methane with the present methanotrophic bacteria in the sand and soil mixes in each reactor, showing average maximum methane oxidation of each individual set of two reactors. These results were obtained during the critical period of approximately two weeks. The average methane consumption of the sets were 11.1, 10.6, and 6.4% for Set 1, Set 2, and Set 3, respectively. Set 3, comprising of two reactors containing soil to sand in a ratio of 1:9, showed different and non-uniform oxidation efficiencies of each one of the two reactors; even though, each reactor was filled with the same mixed material and was subjected to the same internal and external conditions. This non-uniformity was also present in the other two sets of 1:1 and 9:1 soil/sand ratio mixes (Figures 4.10 and 4.9, respectively); however, this fluctuation was seen to be more pronounced for all of the reactors occurring at the first 2–5 d from the start of the experiment, reaching more uniformity toward the end of the experiment. These fluctuations could be due in part to the makeup of the microstructures of the soil and sand mixes in the reactors. Although the soil materials were packed in the same way inside the reactors and with the same mixed ratios, the microstructure of the alignments of the grains of the materials and the nature of the connectivity of the voids within each mix of the

materials varied. (Section E.4, Appendix E). When the experiment was first started, methanotrophic bacteria in the soil and sand mix were not subjected to continuous flow of methane. Therefore, the bacteria were in an inactive state to mitigate methane. Similarly, when the gas mix of oxygen and methane were introduced, the gas had to navigate through the least resistive path in the soil, avoiding the closely connected grain paths and following through into the less restricted voids, therefore, reaching first the group of the bacteria closest to its path. In a short period of time and depending on this soil's microstructure, different interactions could occur; whereas, for a longer period of time, the gas mix would have saturated the microstructure of the soil materials, producing less fluctuation in the results, particularly for low methane loading of 0.8333 ml/min, as evident from these figures. The second reason for these observed fluctuations could be due to the low accuracy of the bubble meters, in which adjusting accurate rates of gases entering or measuring the leaving outputs, using these meters for each reactor could pose a problem. In particular, since the input and output rates of variables were low, small variations in inputs and outputs influenced by even small temperature variations could have sensitive outcome on these oxidation rates. To have accurate gas flow meters for each reactor would have been too expensive for this experiment. In addition to the inaccuracy of the bubble meters, samples of inputs and outputs to be taken for all the six reactors and moving extracted samples for measurement, using the GC analyser were not taken at once. Some time difference was required to process each sample of the several inputs and outputs of each reactor, and which could have little effect on the outcome on the results. For each of these measurements, the GC accuracy comparisons of the results with standard samples showed an accuracy ranging from 95–98%(Figure 3.1). Finally, measuring biological interactions are complex and different from measuring physical parameters, in that, the reaction of each single cell is influenced by each surrounding microenvironment. The overall outcome is the sum of all activities of each one of these bacteria, where small changes can easily affect the total outcome of the reaction. These obtained results in this experiment could again be taken indicative.

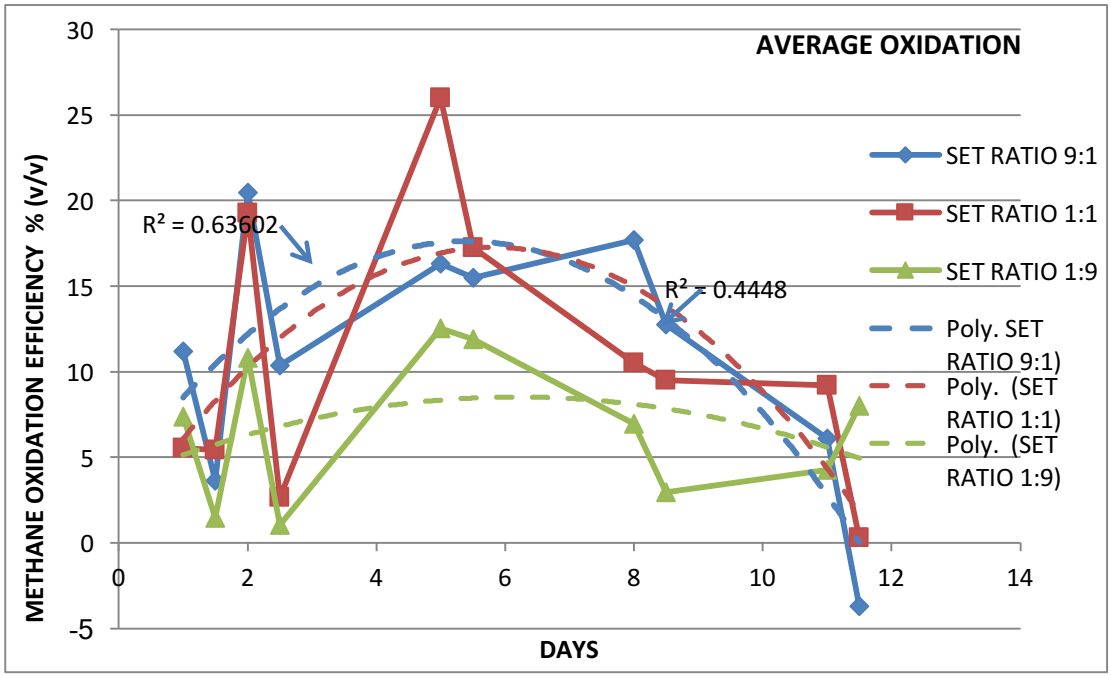


Figure 4.12: Average methane oxidation efficiencies for column reactors sets 1, 2, and 3.

The other observation that can be seen from Figures 4.9, 4.10, and 4.11 is that Set 1, the soil to sand ratio of 9:1, which had a higher soil to sand ratio, exhibited an average oxidation of 11.1%; this was higher than Set 2, which had a 1:1 ratio. In sequence, Set 2 exhibited an average oxidation of 10.6%; this also was higher than Set 3 of a 1:9 ratio, which had an average methane consumption of 6.4%. Noticeably, the higher amount of soil material in Set 1 led to higher amount of OM and MC in the mix, reaching 18.56% (wt/wt) and 26.08% (wt/wt), respectively; while the higher value of sand in the mix in Set 3 led to lower values of 0.67% (wt/wt) and 1.45 (wt/wt) for OM and MC, respectively. Soil materials in the sets were ground soils, exposed to ground bacteria and weather environmental conditions, leading to higher OM and MC; while sand materials were clean standard materials, which have not been exposed to those same conditions. Consequently, the higher soil material mixes were favourable mix conditions for the methanotrophs to function more actively, hence, to achieve higher methane consumption, as indicated in Table 4.2.

The results of the average oxidation rates of each set, shown in Figure 4.12, indicated that the material of lower grain sizes had less void space volume than the more coarse soil to sand ratios. Less void spaces of finer grain sizes support more methanotrophic bacteria than the coarse grain-sized materials (Sections E.4 and E.5, Appendix E). However, coarse material has the property of high porosity that could allow better oxygen diffusion, given that moisture and nutrient conditions are available. For Set 1, the porosity was calculated to be 17.5%, showing a high percentage of small particles in the set vs the porosity of Set 3, evaluated to be 43.7%, thus allowing higher gas diffusion for Set 3 than that of Set 1.

Another observation that can be seen from Figure 4.12 is that, while Set 1 had a finer grain ratio of soil to sand set at 9:1, which exhibited higher average consumption, the average methane consumption decreased faster compared to the coarser material of 1:9 of Set 3, or the balanced grain-sized material of 1:1 with the passing of time. This decrease in oxidation could be attributed to the fast clogging of the soil of the filter due to the formation of EPS produced by the bacteria (Section 4.5), particularly from the bacteria existing in confined and small voids of the reactors. This result could have been attributed to the potential of the coarse material to allow easier gas passage through the material better than the fine-grained materials, and while the fine-grained reactors can have faster clogging of the filter. If oxygen were to penetrate deeper, it would have the potential to oxidise methane with higher efficiency, but in the meantime, it had the potential to clog the soil even faster than the other reactors. This result is in line with the conclusion reached by Dammann et al. (1999) and Streese et al. (2001, 2003, 2005), through conducting a biofilter experiment with gradually varying grain size bed material, and noticing a fast clogging of the fine-grained section against the larger one (Appendix-B, causes of clogging are discussed in Sections 4.3.3 and 4.3.4). The consumption efficiency for Set 1 even went into the negative zone, reaching a rate of - 3.7%, indicating that methane was generated anaerobically, in which more methane was added to the already existing methane within the reactors (Section 4.4 for the EPS formation).

Another observation that can be deduced from this experiment is that the balanced ratio of soil to sand of 1:1 could produce high oxidations when compared to the material of the finer grained soil to sand ratio. Thus, having finer grain soil ratios than this balanced set does not extensively add to the oxidation efficiency, as evident from the average oxidation efficiencies of the three sets (Table 4.3), of which each set had an average oxidation efficiency of 11.1, 10.6, and 6.4%, for fine soil to coarse sand ratios of 9:1, 1:1, and 1:9, respectively.

Finally, methane oxidation efficiencies were observed to be low as the norm in this type of material setup, irrespective of the variation in grain sizes of the soils, to produce higher efficiencies for methane elimination. When methane gas was passed through the soil medium containing methanotrophic bacteria that had been mixed with sand of larger aggregate sizes, the result showed low methane oxidation efficiencies (Figure 4.11). This result has confirmed that these types of soils are not necessarily the most efficient types of material used for landfill covers as a means of eliminating methane. This runs parallel to the results of the batch test carried out in Section 4.1, whereby reactions did not produce appreciable changes when nutrients were insufficient to warrant some activities, as shown in Figure 4.1. Nevertheless, this result is expected in this column experiment, because the materials used were only sand and soil, with low biological contents added to them. Adding nutrients to the batch experiment samples and exposing a larger sample surface to air regenerated the bacteria and led to their aggressive consumption of methane in the batch experiment. If nutrients or amending the reactors with nutrient-rich materials, such as compost, sewage waste, or any biological matter were to be added in this column experiment, it would evidently result in higher reactions of oxidation, in a similar fashion to the systems tried in the existing literature (Tables 2.5, 2.6, and 2.7). However, that would defeat the purpose and the intention of using this type of material as a cover for arid environments as proposed in the first place.

Sand and soil materials used in these column reactors (shown in Figure 3.2) were selected to simulate the reaction of typical materials present in most regions of arid environment for landfill use, particularly in the arid environment of Kuwait, where sand

and soil are abundant and are exclusively used to cover wastes. Therefore, it is important for the choice of materials to provide convenience, favourable environment and to be cost-effective to the landfill operators to warrant their use for efficient oxidation rates. Garden soils, as shown in the batch test results in Section 4.1.2, showed the presence of methanotrophic bacteria that could regenerate and stay active to oxidise methane. In this column setup, the purpose of mixing the garden soil, which contained methanotrophs and mixed with sand (or any larger grained materials, such as crushed glass, shredded plastics, gravels), was to take advantage of the characteristic of sand as having larger grain sizes, which would introduce larger pore sizes in the mix, but with less effective surface area. Consequently, the larger pores could sustain more oxygen in the voids created by these sand particles and would allow the passage of gases within the mixed material to encourage oxidation. In this setup, methane, oxygen, and the soil containing methanotrophs were all present in the column containers, which would provide an opportunity for the bacteria to be active in converting methane gas to carbon dioxide. However, the missing organic matter (OM) from the system had reduced the efficiency process (see Section 4.7).

This experiment when compared with the findings of He's experiment, where only clay soil, having a low OM of 0.54 %(wt/wt) was used, showed that oxidation reached almost the same rate level as that of Set 1, even though the clay soil used in He's experiment had high grains surface area and with low total nitrogen content. This would imply that organic matter is an important additive to be considered when using a material for landfill covers (Section 4.7 on discussion of the effect of organic matter on methane oxidation).

The column experiment conducted at the pilot laboratory at Cassie Building provided important information. Firstly, sand and soil as cover materials cannot sustain oxidation efficiently, unless all favourable methanotrophic microenvironments discussed in Section E.2-E.24 (Appendix E) are provided and maintained. Secondly, for most of the landfills using these types of materials as landfill covers, measurable methane oxidation is not likely to occur within that soil if biological materials were not mixed with them, hence, providing more nutrients and better oxygen diffusion.

Unfortunately, due to environmental concerns, landfill regulations prevent mixing landfill cover soils with biological matters (Eureopean Directive 1999/31/EC, 1999). When using this soil and sand material as a landfill cover, as it is the norm practiced in countries with a desert environment (among other countries for different reasons), the cover will not sustain methanotrophic oxidation fully, as the biological matters are scarce, and as dust fallout will likewise clog the survace voids of the cover, hence hindering the full potential of such covers. Therefore, this would necessitate finding some other kind of mechanisms to provide oxygen deeper down to the oxidation layer where methane and biological matters are all present.

4.3 Effects of oxygen penetration inside the soil on methane oxidation

Soil of specific characteristics indicated in Table 4.4 was added to a compost material and investigated using continuous flow reactors. The characteristics of this loamy sand soil are pH= 5.8, average porosity= 47.5%, OM= 45.46% (wt/wt), and MC= 30.53% (wt/wt). The bulk material of this soil was placed in a container, as one stock material, divided, and used in each of the three sets of this experiment (Section 3.4.4).

Figure 4.13 illustrates the outflow data obtained from operating the three sets of reactors of Figures 3.3 and 3.5, that lasted for 35 d to investigate the effect of providing oxygen deep inside a soil on the methane elimination. The data presented in this figure and in following Figures 4.14, 4.15, and 4.16, show that methane oxidations were approximately uniform and of the same range for the six reactors during the first 14–15 d of operation; however, from that point on, the data became divergent for each set with some fluctuation. It could also be observed that the concentration decreased continuously for all of the reactors for the first four days since the start of the experiment, indicating that the systems were at their transient states. Moreover, they started to increase continuously from that point on for another ten days and for all of the reactors. This oxidation behaviour of the bacteria in the soil mix for the first few days is expected, since the low rate of methane (1 ml/min) and the diffusion time required for both, the oxygen and the methane, to reach the bacterial community was slow to saturate the soil structure. This is also in line with the result obtained from the

literature as indicated in Figure E.7 (Appendix E). When the system reached a steady state, and with sufficient time for the gases to infiltrate into the voids of the soils, the micro-environmental conditions engulfing bacteria communities started to have an effect on oxidation. Set 3, where the physical port of the air supply was located deep down at a 26-cm depth below the surface of the soil in the reactors, and where a maximum amount of air to saturate the soil mix was supplied, showed a clear higher oxidation efficiency than the other two sets. The final oxidation efficiency of methane reached 42.503% v/v for Set 3 at the end of the experiment, compared to 25.133% and 2.385% for Sets 1, and 2, respectively; yet with maximum oxidation efficiencies of approximately 49, 56, and 58% for Sets, 1, 2, and 3. Results would infer that the higher retention time of the oxygen with methane in the soil for Set 3 allowed the methanotrophic bacteria to assimilate methane with higher efficiency, resulting in a profound effect on the methane consumption. When oxygen was introduced in Set 3 at a -26 cm from the surface of the soil, it mixed with the methane gas that was introduced from the bottom of the reactor, allowing the oxidation zone to widen substantially, therefore supplying wider bacterial communities with their needed methane and oxygen gases for more interactions. This wider zone of interactions is analogous to the schematic representation shown in Figure E.5 (Appendix E); which oxygen and methane run contrary to each other, allowing smaller oxidation zones for interactions as that of Sets 1 and 2 (Section E.11, Appendix E). Table 4.5 shows the input parameters to the reactors, along with the average oxidation efficiencies for each reactor and for each set; whereas, Table 4.6 shows the loading and efficiency capacities of each set and of each reactor. By using all of the data collected during the course of the experiment, the average oxidation concentrations were 21.53%, 34.17%, and 35.66% for Sets 1, 2, and 3, respectively, as indicated correspondingly in Figures 4.14, 4.15, and 4.16. These efficiencies showed that oxidation was better for the oxygen delivered deeper into the lower layer than the shallower ones, by as much as 65.7% at the end of the experiment. In this setup, it is interesting to note that Set 1 represented a system of air supply that was just touching the surface of the soil, simulating a landfill cover with air that interacts only with the soil at the surface of the cover. This representation, indicated by Set 1, is the basic representation that all designed systems of methane assimilation relied upon (Appendix A). Additionally, at the end of this

experiment, the average oxidation efficiency between Set 3 and Set 1 reached 20-folds higher for Set 3 over the efficiency reached by Set 1. This was evident from the plot of the average oxidation efficiencies of all of the sets, as exhibited in Figure 4.17.

Exp.	Soils classification	Porosity (%)	pH	Organic Matter (OM) (%) (wt/wt)	Moisture Content(MC) (%) (wt/wt)
Sets 1, 2, 3	Compost, loamy sand (0–2 µm: 4%, 2 µm–63 µm: 14%, 63 µm–2 µm: 82)	58.2	5.8	45.46	30.53

Table 4.4: Characteristics of the soil used in the experimental sets (values are the average of two reactors for Sets 1,2, and 3)

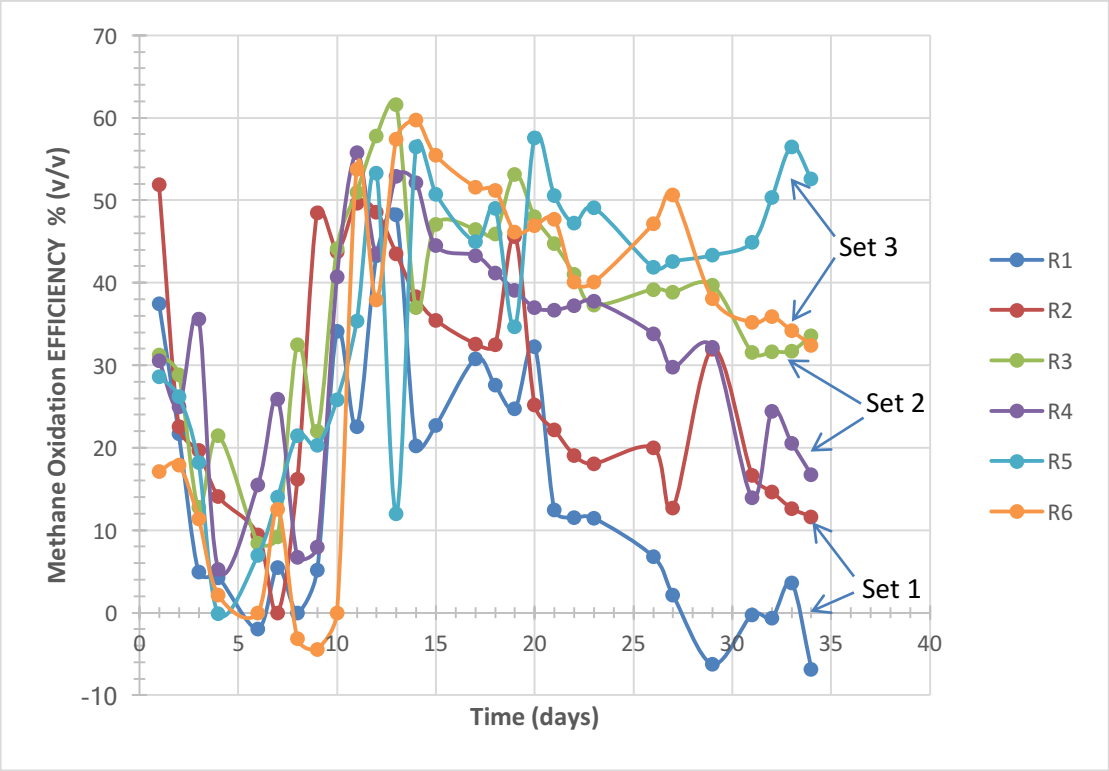


Figure 4.13: Methane oxidation efficiencies for system of three sets of reactors, with air introduced at different sections.

For all of the investigated sets in this experiment, the oxidation rates tended reduce over time; even though, the supply of oxygen and air was kept at the same amount and at the same constant levels. This behaviour could be due the formation of the EPS formed along the preferred path travelled by the gas molecules, reducing the interaction between the bacteria and these molecules when the slime-like material (polysaccharides) inhibited the transition of the nutrients to the bacteria. This is evident from the published literature shown in Figure 2.3 and from the following sections 4.4 and 4.5. In addition, this behaviour was seen to be associated mostly with the reactors of a lab experiment, since the interactions were limited to small confined spaces of the reactors; while field testing showed little evidence of the formation of EPS in cover soils, as noted by Huber-Humer, 2005.

Sets (level)	**Reactor**	**Methane Input (ml/min)**	**Air (oxygen) Input (ml/min)**	**Average Reactor's Methane Oxidation (%v/v)**	**Average Methane Oxidation (%v/v)**
Set 1 (0 cm)	Column R1	1.0	3.3333 (0.6666)	15.70	21.53
	Column R2			27.36	
Set 2 (-13 cm)	Column R3			36.70	34.17
	Column R4			31.63	
Set 3 (-26 cm)	Column R5			36.93	35.66
	Column R6			34.66	

Table 4.5: Input parameter and average oxidation efficiencies for the column reactor sets.

To establish a comparative analysis between the findings of these experiments and the findings in the published literature, results of comparable experiments obtained by

Scheutz (2005) are shown in Table 4.6, along with the maximum oxidation rate and the maximum efficiencies obtained for all the six reactors of this study. These maximum oxidation values ranged from 162.2/167.0- g CH_4 $m^{-2}d^{-}$ for Set 1 to 189.7/200.8- g CH_4 $m^{-2}d^{-1}$ for Set 3; while Set 2 indicated a maximum in the same range as that of Set 3, equal to 187.0/194.1- g $CH_4m^{-2}d^{-1}$. The efficiencies of these sets were 48.3-49.7%, 55.7-57.8%, and 56.5/59.8% for sets 1, 2, and 3, respectively. This shows a 10% difference between Sets 1 and 3 at maximum efficiencies, indicating the previously established fact that supplying bacterial community with their needed oxygen deeper inside the soil would be substantially more advantageous than relying only on the surface diffusion mechanism of supplying oxygen. However, this difference became even much wider, as the time of the experiment went forward, which indicated a difference in efficiency exceeding more than 65% at the end of the experiments as aforementioned.

Set No.	**Reactor No.**	**CH_4 Loading ($g\ CH_4\ d^{-1}m^{-2}$)**	**Max. Oxidation rate ($g\ CH_4 d^{-1}m^{-2}$)**	**Efficiency (%)**	**Temperature (^oC)**
1 (0 cm)	R_1	335.8	162.2	48.3	22+/-2
	R_2		167.0	49.7	
2 (-13 cm)	R_3		194.1	57.8	
	R_4		187.0	55.7	
3 (-26 cm)	R_5		189.7	56.5	
	R_6		200.8	59.8	
Compost/Sand[a] (1:1) (Scheutz et al., 2009b)		229–254	161	48	22
Compost/Sand (1:5) (Scheutz et al., 2009b)			29	12	

[a] Rich in organic matter.

Table 4.6: Loading capacities and oxidation efficiencies of sets, 1, 2, and 3, in comparison with surface diffusion experiments.

Scheutz et al. (2009b) performed experiments on compost material, which was rich in organic matter and mixed with sand. Their findings indicated that when methane loadings of 229–254 **$g\ CH_4\ d^{-1}m^{-2}$** were introduced in column experiments, oxidation rates

of 161 and 29- g $CH_4 m^{-2} d^{-1}$ for a compost material mixed with sand in a ratio of 1:1 and 1:5, respectively, were realised. These findings indicated that the oxidation efficiencies of these mixes were 48% and only 12% for compost/sand ratios of 1:1 and 1:5, respectively. The findings of the compost mix of 1:1 can only be compared with the findings obtained for Set 3 of the experiment described in this section, since both have the same mix ratios, and both relied on oxygen being supplied all along the depth of the cylinders. Set 3 mixing ratio was 1:1 of compost and soil with granular makeup described in Table 4.4. In these two sets, the maximum oxidation rates of 189.7/200.8 $gCH_4\ m^{-2} d^{-1}$ (Set 3), for methane loadings of 335.8 $gCH_4\ m^{-2} d^{-1}$, were compared with 161- g $CH_4\ m^{-2} d^{-1}$ of Scheutz's experiment; while the efficiencies reached 56.5/59.8% for Set 3, and 48% for the respective experiment of Scheutz's were all values of close proximity. These differences between the experimental results are expected however, since characteristics of compost materials differ substantially from one vender to another, due to the lack of characterisation standards, unlike the well-established standards used for sands, for example. Compost materials could differ in their original structural makeup, maturity rates, and granular characteristics, in addition to other issue associated with using compost material in mitigating methane, which all make the outcome of using compost material to some extent, unpredictable (Section 2.6).

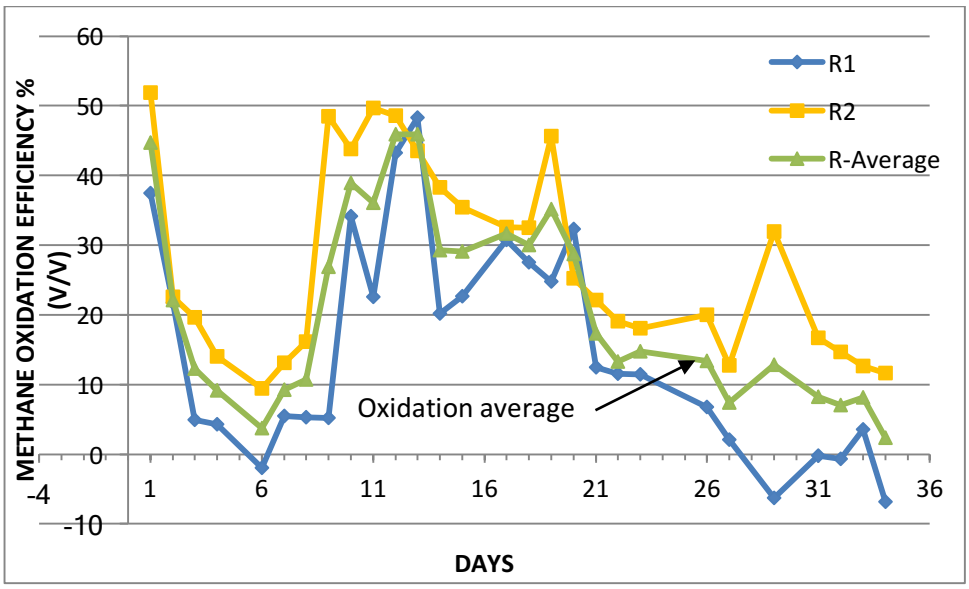


Figure 4.14: Methane oxidation efficiency for set 1.

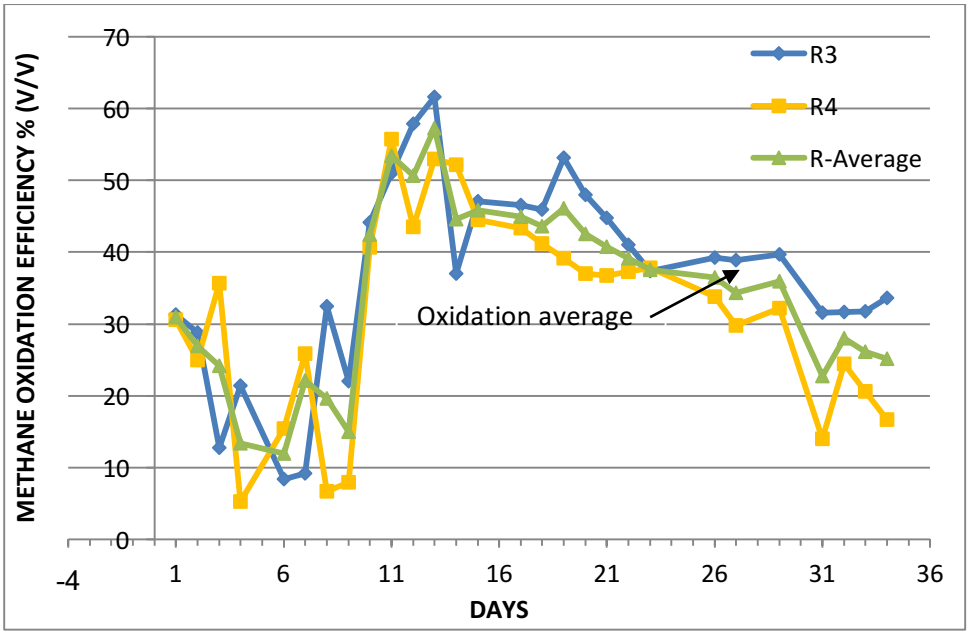


Figure 4.15: Methane oxidation efficiency for set 2.

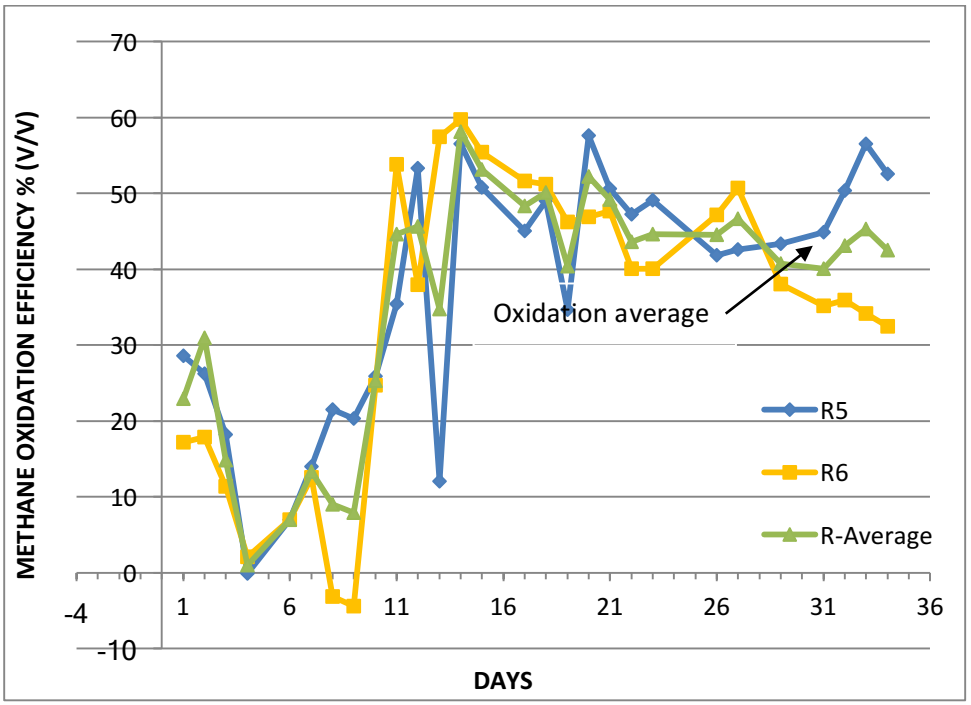


Figure 4.16: Methane oxidation efficiency for set 3.

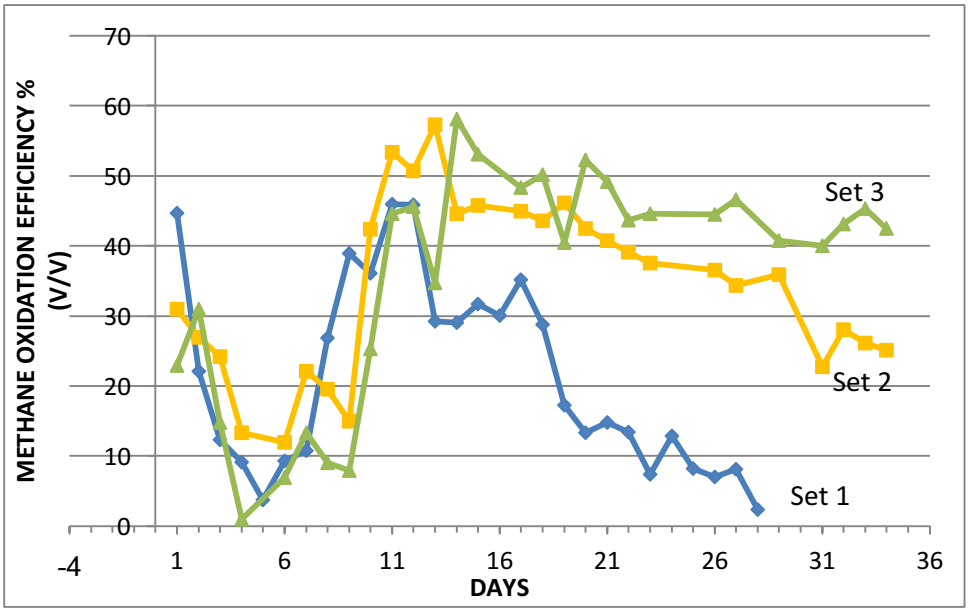


Figure 4.17: Average methane oxidation efficiencies for sets 1, 2, and 3.

From the result of this experiment, it was clear that the efficiency of delivering oxygen deep inside a cover soil was much higher than the ones that relied on oxygen supply to only the surface of the soil, as was anticipated. This efficiency that reached higher than 65% at the conclusion of the experiment (Figure 4.13), could reach even higher levels, if the delivery mechanisms were to be extended even deeper still into the lower levels, further extending the oxidation zone. This would be useful in building a passive barrier system for mitigating landfill gas emissions for arid environment, having hindered air diffusion at the surface by particulate matters.

4.4 Methane mass balance, porosity, and the formation of EPS

Formation of EPS in column and in the field soils is a well-known occurrence to the research community (Costerton et al., 1878; Hilger et al., 2000; Chiemchaisri et al., 2001; Streese and Stegmann, 2003; Wilshusen et al., 2004b; Marvasi et al., 2010; Lee, et al., 2015). In actuality, EPS formation is common for most bacteria and essential for their own survival in their natural environment (Davey and O'Toole, 2000). This extra cellular substance is formed as a bio-film by the bacterium itself in a matrix encapsulating its body, doing so for the purpose of protecting against other bacteria's

predation, preventing cell desiccation against extreme conditions, and streamlining vital elements for transport to the inside of the cell. EPS is an amorphous slime or a polymer gel bio-film made of polysaccharides, consisting mainly of sugars (37–36%) and amino acids (30–38%), designed to hold bacterial community together (Hilger et al., 2000). Cells draw nutrients by inducing an osmotic gradient through this varying thickness outer shell EPS (Marvasi et al., 2010).

To understand the reason behind methane uptake deficiencies as time progressed in the column test filters conducted in this research, series of tests must be taken, as suggested by Hilger et al. (2000). These tests included filtration rate, glucose assays, total carbon, and methane oxidation potential. To perform these tests again would be a duplication of efforts, time, and financial resources. Nevertheless, results obtained in the literature can be discussed and compared with the outcome of the tests conducted in this study, to demonstrate the effects of EPS formation on methane consumption efficiencies.

Hilger et al. (2000) used sandy loam soil collected from a closed landfill cover in four column reactors. All were prepared for investigating EPS formation, when all bio-filters were subjected to the same conditions. The four reactors were injected with synthetic gas containing methane and carbon dioxide, having a mix of 50/50% at a rate of $3.25x10^{-7}$ g CH_4 cm^{-2} s^{-1} (10 cm^3/min). The columns were injected with flow of air supplied on the top soil surfaces of each of the four reactors at a rate of 50 cm^3 min^{-1}. The results of these tests, that were conducted by Hilger and others were averaged out from Figure 2.3 for the four reactors, and plotted along with the averaged results of the duplicate samples obtained for Sets 1, 2, and 3 (Figure 4.13) for comparison. These averaged results are presented in Figure 4.18, showing operation time of 34 days and approximately 900 hours for sets 1, 2, 3 and Hilger's, respectively..

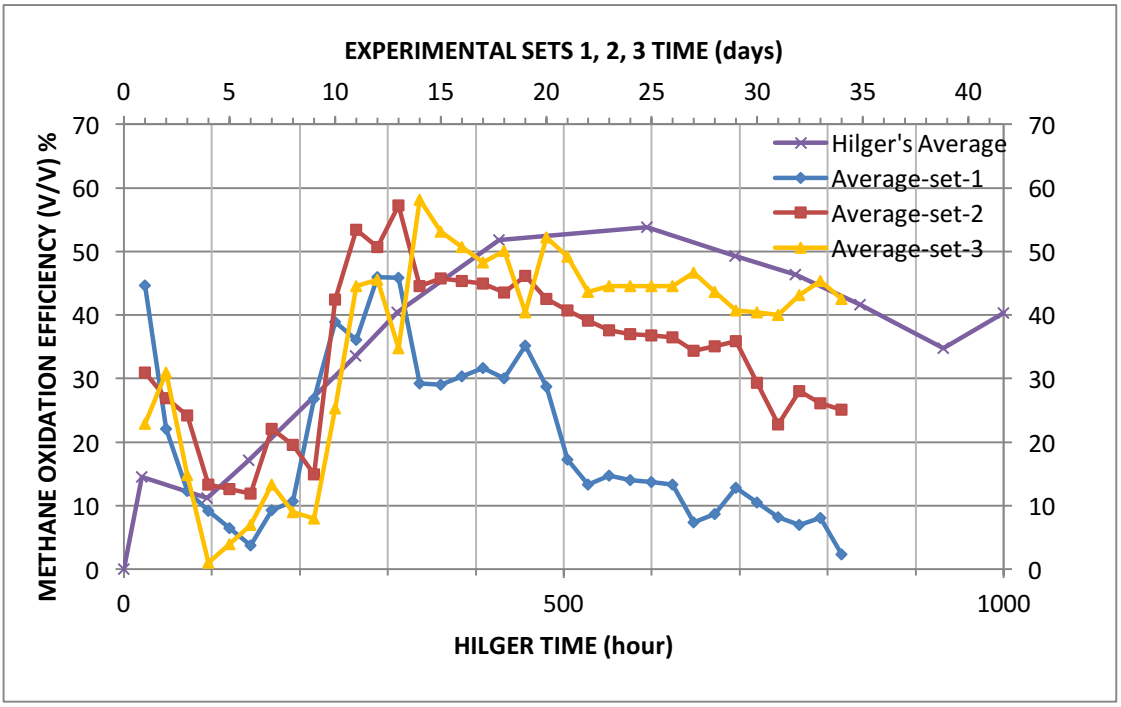


Figure 4.18: Comparison of oxidation efficiencies between the results of Sets 1, 2, and 3 and with results obtained by Hilger et al. (2000).

The results in the figure, showing methane consumption for Sets 1, 2, 3 and of Hilger's experiment are all increasing until reaching a maximum plateau, then decreasing continuously with steady rates of decline. The increase of methane consumption for Sets 1, 2, and 3 were steady at an approximate rate of 17.5% (v/v) day^{-1}, starting after 6 d of operation; while Hilger's oxidation outcome was at slower consumption rate of 3.33% (v/v) day^{-1}, commencing after 5 d of operation. The consumption percentage peaked at approximately between 45 and 58% (v/v) for Sets 1, 2, and 3, after 11–13 d of running time; while in Hilger's case, the consumption peaked at 51% (v/v) after 23 d. The decline rates after peak were 1.674, 1.136, and 0.565 % (v/v) day^{-1} for Sets 1, 2, and 3, respectively, compared with a rate of 1.067 %(v/v) day^{-1} for Hilger's experimental results. It is clear from these declining rates for the three sets that they were caused by the supply of air at different levels, as previously noted (Table 4.7). For Set 1, oxygen supply was injected at surface, allowing only limited amount of oxygen to penetrate at that level. This supply had caused the highest decline; while Set 3, where oxygen supply was delivered at level -26 cm below the surface of the reactor, the supply had caused the lowest. The decline rate of Set 2 was at an intermediate level, producing a rate of

between the other two sets. In comparison, air input of 50 ml min^{-1}, pumped over the surface of the soil and a 50/50 methane/carbon dioxide input of 10 ml min^{-1} injected from the bottom had been used in Hilger's experiment, compared to 3.333 ml min^{-1} and 1 ml min^{-1} for air and methane, respectively for Sets 1,2, and 3 experiments. Approximately fifteen and five times for air and methane, respectively, were used in Hilger's case more than the air and methane that were used in Sets 1, 2, and 3. These high rates had caused longer oxidation time for Hilger's case, but eventually reaching almost the same level of methane removal as for the three Sets 1, 2, and 3. However, both rates of removal in Hilger's experiment and of Set 3, had declined by almost the same levels, after reaching the same maximum oxidation. This could be due to the high quantities of gases inputted for both, as indicated in Table 4.7. This table shows the operating conditions for all the three sets and for Hilger's experiment, for comparison purposes. Obviously, the higher volumes of oxygen and methane supplies in the case of Hilger's experiment delivering more needed gases (methane and oxygen) to the bacterial communities caused longer oxidation rates. However, after 11 and 21 d of running time both for Sets 1, 2, 3, and Hilger's, respectively, have experienced steady decline, which, as yet needs to be explained.

One reason for these declines in oxidation efficiencies, as indicated in Figure 4.18, was the formation of the EPS, predicted by the reaction equations 2.1 and 2.2, in which the bacteria exerted substantial biomass. This biomass could have clogged the pore spaces existing between the grains of the soil, hence, hindering the transport of the needed gases to the bacteria.

Conducting a series of tests, Hilger and group have confirmed the already known fact that EPS formulation was indeed responsible for reducing methane oxidation, as they found a dense polymers material, coating the grains of the bed material with clumps of polysaccharides. The clumps were apparent when treating the materials with a series of chemical and physical tests, then subjected to a blue die so that they can be viewed through a microscope, as they appear in Figure 4.19. These clumps engulfed the dead bodies of the cells forming these substances, as shall be seen from detailed studies later in this section.

Experi-ment	Filter Material	Gas Input (ml min^{-1})	Air Input (ml min^{-1})	Air level cm	Organic Matter (OM) (%) (wt/wt)	Total Nitrogen (TN) (%) (wt/wt)	Moisture Content (MC) (%) (wt/wt)	Total Carbon (TC) (%) (wt/wt)	C/N Ratio	Run Time
Set 1	Compost/soil 1:1	1 CH_4	3.333	0	45.46	1.53	30.53	29.62	18.95	34 d
Set 2				-13						
Set 3				-26						
Hilger et al., 2000	Sandy loam	10 CH_4/CO_2	50	0	27.0	0.025–0.03	15.0+/-5	0.32–0.66	13–22	242–2808 h

Table 4.7: Comparative data of the sets 1, 2, and 3 with the data produced by Hilger et al.,(2000) [(data for Sets 1, 2, and 3 are average of two reactors)]

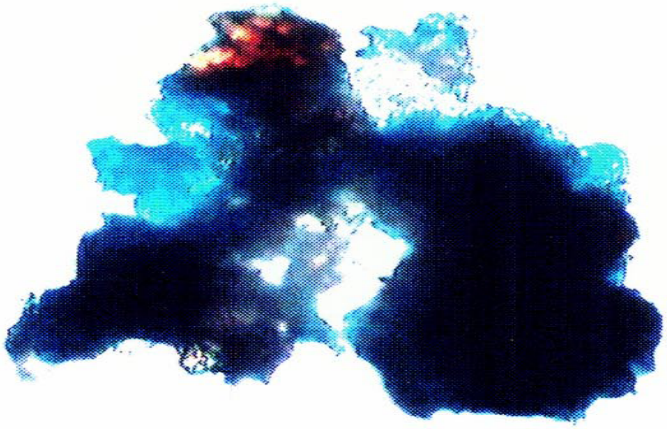


Figure 4.19: Exopolymeric substance (EPS) as seen through a microscope (source: Hilger et al., 2000).

To test the clogging, a mass balance between the input gases and the output of materials and gases must be calculated. When methane is consumed by the bacteria, in the presence of oxygen and nitrogen (in the form of ammonium), a biomass ($C_4H_8O_2N$) is formed along with other gases in an amount depending on the bacterial types present in that particular soil space of the column reactors and on the type of pathways used by the bacteria. A biomass of 12.043- g $C_4H_8O_2N$ mole $^{-1}$ (0.118 X102.113 g mole^{-1}), or 10.416- g $C_4H_8O_2N$ mole^{-1} (0.102 X102.113 g mole^{-1}) can be produced for ribulose or serine pathways, respectively, when a mass of 16.043 of CH_4 is consumed using the molecular weight of elements (C: 12.0107, O: 16, N: 14.0067, H: 1.00794 g mole^{-1}), in accordance with equations 2.1 and 2.2. To arrive at the weight of biomasses accumulated, causing the blockage and the formation of the EPS during the course of the experiments, it was required that the total masses of methane consumed to form these biomass must be calculated. Table 4.8 presents the average results of the mass of methane consumed for the duplicate reactors of Sets 1, 2, and 3. Also, the table shows the average mass of the same for the four reactors in Hilger's experiment, for the same duration of only 34 d for both of the two experiments, for reason of comparison. Total masses of 6.68, 11.13, and 12.26 g of methane were consumed by the bacteria during the course of the experiments for Sets 1, 2, and 3, respectively, compared with a total mass of 64.38 g consumed by the bacteria in Hilger's experiment, for the same period of time. Results exhibited an increase in mass consumption for Sets 1, 2, and 3, due to the amount of methane exposure and to the level of oxygen introduced inside the reactors. It also indicates the formation amount of biomass due to this methane assimilation in accordance with the reaction equations described in Chapter II, at only 5.02, 8.36, and 9.21 g for the experimental Sets 1, 2, and 3, respectively, compared to 48.35 g of $C_4H_8O_2N$ in Hilger's experiment and all for the ribulose pathway. On the other hand, for serine pathway, the biomasses formed were 4.34, 7.27, and 7.96 g for Sets 1, 2, and 3, respectively, compared to 45.07 g for Hilger's experiment. These results show much higher biomass accumulation in Hilger's experiment than the accumulations produced by Sets 1, 2, and 3, which could be attributed to the high methane input of 10 ml/min of ratio 50/50 CO_2/CH_4 in Hilger's case, vs. only a 1ml/min input for Sets 1, 2, and 3. However, this high accumulation of mass in Hilger's experiment is distributed over a much larger volume space, which

brings the density to comparable levels with the densities of Sets 1, 2, and 3, as shall be seen from following analysis.

Bacterial group I (and sometimes group X) uses the ribulose reactive pathway; while bacterial groups II and X use the serine reactive pathway. Unfortunately, little information exists on the distribution profiles of each of the bacterial flora along the depth of the bio-filters, at any given time, and at any given methane, oxygen and nitrogen concentrations, so that a known pathway can be used for the calculation of the biomasses. Figure 4.22 suggests that the two types of bacteria, type I (including type X), and type II are present all along the depth of the bioreactors with varying concentrations. Yet still, the distribution of each type along the depth for any given time is difficult to measure. Therefore, calculating an exact biomass distribution as a result of the methane consumption, given this kind of difficulty, would not be possible. Nevertheless, even distribution can be assumed of the two groups of bacteria (hence, even type of pathways) being present, and an approximation would then be possible. This approximation could give an indication as to how much of the biomass would be accumulated, and how much of that would affect oxidation rates. The average of the biomass produced due to the rubulose and serine pathways are calculated in Table 4.8, showing indicative values of 4.68, 7.79, and 8.58 g of $C_4H_8O_2N$ for Sets 1, 2 and 3, respectively and 45.07 g for Hilger's. It is interesting to note that, while the biomass accumulation of Set 1, being only of average 4.68 g compared to Set 3, with an average 8.58 g of biomass, the rate of decline after peak for Set 1 was higher at 1.67% day^{-1} compared to 0.57% day^{-1} for Set 3. The reason for this lower biomass/higher decline could be attributed to the fact that Set 1 has oxygen fed at the surface of the soil, diffusing only to low levels of the soil's surface. This shallow oxygen penetration allowed the bacteria to accumulate a higher concentration level of total biomass at that level of the top soil. However, the oxygen penetration was limited by the fast accumulating EPS, and likewise, limiting further oxygen and nitrogen supplies to be delivered to the bacteria at the lower levels. In contrast, the wider oxygen distribution for Set 3, which allowed higher biomass accumulation, wider bacterial communities' involvements, and more time for EPS to accumulate, produced low rates of decline in methane consumption. While for Hilger's, an average biomass of 45.07 g was

accumulated, equivalent to 10 times as much biomass of Set 1, and approximately five times as much as the average biomass accumulated for Set 3, having the same decline rates of oxidation with Set 3, likely to be attributed to the much higher rates of methane and oxygen inputs level than the level used in the sets studied in this research.

Sets	Total CH_4 Mass Consumed kg (10^{-2})	Total Biomass Formed (Ribulose) kg (10^{-2})	Total Biomass Formed (Serine) kg (10^{-2})	Average Total Biomass kg (10^{-2})	Density of Average Biomass g/cm^3 (10^{-3})
Set 1	0.668	0.502	0.434	0.468	6.36
Set 2	1.113	0.836	0.722	0.779	10.60
Set 3	1.226	0.921	0.796	0.858	11.68
Hilger's	6.438	4.835	4.179	4.507	8.39

Table 4.8: Methane mass consumption and biomass formation

The biomass produced by the bacteria in accordance with equations 2.1 and 2.2 may not all go into EPS formation. Part of this produced biomass could be used into the cell's own body building, which eventually at some elapsed times could enter into the EPS as dead bacterial debris when the bacteria die. In addition, other bacterial communities that do not depend on methane for their own survival may also exert some other forms of polymeric substances that could add to this formation (Marvasi, et al., 2010). How much of biomass goes into body building and how much enter into the EPS formation are hard to find, since there is the need for a biological balance as well as mass balance. These specific biological calculations are difficult to find, since the biological makeup changes continuously, as a consequence of the changing microenvironment around the microorganisms existing in the soil. Hence, the mass balance discussed in this section could only be estimates of EPS formation due to bacterial activities.

The porosity of the biofilter beds could be affected directly if the density of soil is increased. Examining the densities of the biomass formed at the end of the experiments revealed that the volume of the materials in each of the reactors constituted 735 cm^3, producing densities of biomasses averaged at $6.36x10^{-3}$, $10.60x10^{-3}$, and $11.68x10^{-3}$ g cm^{-3} for Sets 1, 2, and 3, respectively (Table 4.8). These densities signified uniformly varying biomass values accumulated across the three sets, in accordance with the varying amount of oxygen available and the distribution of the bacterial community. Consequently, these densities may not show the total picture clearly, if this lone parameter were to be considered. In the case of Set 1, the oxygen exposure was to the surface of the soil in the reactors, which had induced the formation of the biomass on that surface and in the immediate low level of the surface, creating the most biomass in that level, in response to the limited oxygen available in that same level. While, the rest of the body of the reactive material had little exposure to oxygen, in effect, less or no biomass was formed in the bulk material. The same reasoning could apply to the rest of the sets. Therefore, these densities are not uniformly distributed across the depth of the reactive columns, as they had first appeared to be. The density of the biomass formed in Hilger's experiment, as material of volume 5372 cm^3 was used for each of the reactors, revealed a low density of 8.39 $x10^{-3}$ g cm^{-3}, despite the high mass accumulation. While this biomass showed to be varying or not uniform across the depth; albeit, Sets 1, 2, and 3 and that of Hilger's have added more biomass medium to the bed materials of the reactors.

Considering that the increase in densities due to the added biomasses should have decreased the porosities of the medium and since the density in Hilger's experiment was lower than the densities of Set 2, and 3, the oxidation efficiency of methane should have been more in Hilger's case than that indicated in Figure 4.18. Methane oxidation efficiency for Sets 1, 2, and 3 did not exceed more than 52–58%, and the oxidation was lower than that for Hilger's experiment at 52%. Concurrently, the rate of decline after peak for Hilger's was almost the same as Set 3, even with the added biomass that increased the density of Set 3. Markedly, this would mean that it did not affect porosity; neither did it cause the clogging of bioreactors in the first place. The calculated average porosity of the material initially used in Sets 1, 2, and 3, as indicated

in Table 4.4, was 58%. In contrast, the average of porosities of two reactors at the input horizon, at the close of the experiments, were approximately 53, 48, and 46%, for Sets 1, 2, and 3, respectively, which were of the same order of magnitude as the initial porosity. These porosities changes could be due to secreted materials of the methanotrophic and other bacteria. However, to find out if these changes in the porosities were the cause of the observed high decline in oxidation efficiencies, further investigation was needed.

The indication that porosities change, due to the formation of EPS, may hinder the flow rates of flowing fluids inside the soil medium was tested by Hilger and her group. They passed methane gas again through the four reactors that had been ran for 2400–3300 h (100-137 days), and compared methane migration through them with a control set of reactors of no exposure to methane, thus without EPS formations. The supplied methane in both sets went for 22–26 h, testing to determine whether methane gas would break through in the soil, and of the same rates for both sets, and whether EPS build up would hinder the passage of gas through the bed material of the sets. A noticeable conformity in the results of the two tests was revealed, indicative of no hindrance of methane migration through the soil beds of both sets, despite the formation of the EPS that had been accumulated in the first set. Also, it suggested that gas conductivity and porosity did not affect the passing of methane even after the long period of operation (Figure 4.20). Furthermore, increase in the densities due to biomass build-up (Table 4.8) was in the range of some milligrams per centimetre, consequently, insignificant to have caused substantial changes to the conductivities, as well as to the persistent decline in oxidation rates. Therefore, Hilger et al. (2000) was able to rule out that changes in the porosities in the soil due to EPS formation could disrupt the supply of methane or oxygen to the bacterial communities residing in the soil.

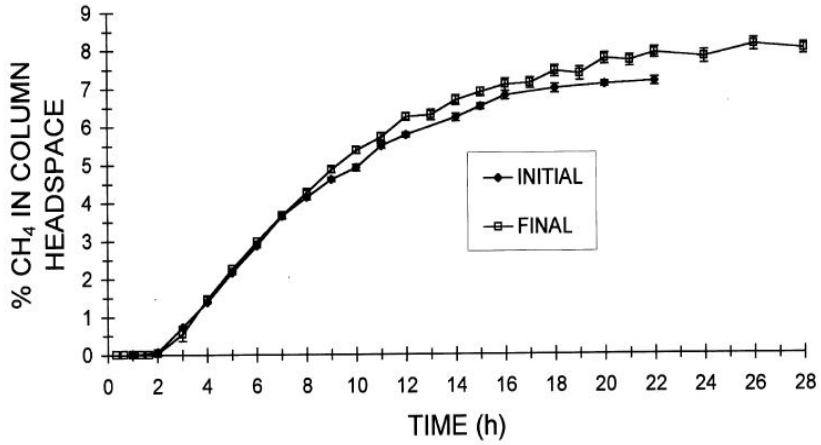


Figure 4.20: Methane migration through soil columns, one tested after a run of 2400–3300 h (final), compared with a second column tested without methane exposure (initial) (source: Hilger et al., 2000).

It was reasoned that the decline in methane oxidation, causing a decline in the efficiency of bioreactors was due to the inability of methane and oxygen to diffuse through the formed EPS bio-film around the bacterium, as it thickens, preventing the gases from reaching the imbedded body of the cells (Marvasi, et al., 2010). The thick EPS material exerted by the bacteria, engulfing the cells, disrupts the osmotic mechanism that these bacteria depended upon for the supplies of the oxygen, methane, and nutrient, thereby leading to the death of the bacteria. This evidence would suggest that diffusivity through the medium of EPS decreases with the increase of the thickness and the age of the bio-film (Marvasi, et al., 2010; Matson and Characklis, 1976). In this process, the bacteria go through four distinctive stages, namely, cells growing aerobically, cells growing fermentatively, cells being dormant and finally, dead cells (Marvasi, et al., 2010; Rani et al, 2007). The decaying dead cells, thereafter, would cause the release of soluble organic carbon and nutrients from the decaying cells. This process encourages the growth of heterotrophic bacteria, consequently competing with the remaining methanotrophs for oxygen and nutrients. Therefore, the EPS formation and the biological cycles occurring in the soil by themselves could neither be held responsible for obstructing the passing of gases through the pores spaces of the soil, nor for affecting the gas conductivities (Hilger, et

al., 2000), even with the addition of dead bacterial debris to the medium soil; but rather could be pointed to the disruption of the transportation mechanisms by the natural formation of EPS around each individual cell, from which the bacteria depended on for the supply of their nutrients, to blame. In addition, the competition among bacterial communities on the available oxygen and the produced dead bodies and debris may be pointed out to have contributed to the deficiencies of methane oxidation. This process is commonly called EPS clogging of the bioreactors. However, it would be more appropriate to call it the EPS blocking of the osmotic transfer of nutrients and gases to the bacterial cells.

4.5 Moisture contents, nutrients effects and causes of EPS formation

While the production of EPS is a natural process used by the bacteria for their own protection, likewise, to serve as a mechanism of anchorage, shield against predators, and a mean to keep the population together; albeit, these bacterial communities are prone to experience stress. When subjected to attacks by antibiotic molecules produced by other microorganisms, ultraviolate radiation, osmotic stress, or surrounded by environmental fluctuations (deprivation of nutrients and nutrients in the form of organic ammonium, high/low rates of moisture contents, deficiency in oxygen, inefficient supply of methane, temperature variations, high and low acidity/alkalinity of the soil, etc.), which all are typical environmental conditions that could happen at any point in time, the bacteria secrete more of EPS's as a natural survival mechanism. However, it is difficult to predict which among these environmental stress-causing factors first trigger this extra secretion of the EPS. One or more of these factors may be working all at the same time. For example, when the bacteria are attacked by another species of bacteria, the bacteria may start the production of extra EPS as a defensive mechanism, which could thicken the biofilm surrounding the cells, hence triggering the inability of the bacteria to draw more nutrients. This action could consequently place further stress on the bacteria, thereby making them excrete more EPS, ultimately leading to their death. This scenario and similar scenarios of other limiting factors may explain the rapid decline in oxidation efficiencies in Sets 1, 2, and 3 (Figure 4.13) and in that of Hilger's experiment and it could explain the difference in results published in the

literature as to the optimum values in enhancing methane oxidation (Table 2.9). To cite varying results of studies, indicating optimal moisture contents for optimum oxidation, Bender and Conrad (1995) suggested 20–35 %wt/wt; Boecks et al., (1996) pointed out 15.6–18.8% wt/wt as the optimum moisture conditions; Einola et al. (2007) found the optimum to be at a range of 21–28% wt/wt. These varying levels of optimal moisture conditions are due to the different granular makeup of the tested soils holding varying amount of moisture, albeit, they are within the range of 15-40% (wt/wt) suggested by Scheutz and Kjeldesen, 2004, in their extensive study over the effect of moisture content on oxidations. Table 4.9 shows the types of materials and the optimal moisture contents that could simulate methane oxidation. These optimal values are comparable to the moisture contents maintained in Sets 1, 2, and 3 experiments, which ranged between 29.60 and 31.46 %wt/wt, and that of the experimental procedure performed by Hilger et al. (2000), which was in the range of 15 +/- 5% (wt/wt) (Section E.13, Appendix E). Therefore, moisture deprivation may not have been the cause of instigating the formation of EPS in the experimental Sets 1, 2, and 3.

Methanotrophic bacteria assimilate organic nitrogen compounds, ammonia, and nitrite as a source of nitrogen. However, the availability of these compounds to enhance oxidation and their concentrations in the soils has become popular topics for debate among researchers. Section E.14 (Appendix E) presented the limitations and conclusions, and the limiting values were indicated in Table E.5, showing diverging views on how each of these added compounds could simulate or inhibit oxidation. Wilshusen et al. (2004b) pointed out that EPS is produced by type I methanotrophs as a carbon cycling process in the event of limited nitrogen in the soil. When there is limited nitrogen, methanotrophs type II flourish in this limited nitrogen environment, caused by the obstruction in the formation of EPS, and can fixate nitrogen from the air directly and oxidise methane at a low oxygen supply. Methanotrophic bacteria can only assimilate methane when there is enough nitrogen in the soil and in the amount dictated by equations 2.1 and 2.2, but with limitation that the ratio of carbon content in the soil in relation to nitrogen (C/N) must not exceed 10, as discussed in Section E.14

(Appendiix E). This deciding factor is important, especially for arid land climates, where rich nitrogen organic matters are scarce.

The two sets of experiments conducted in this study showed that C/N ratios were seen to be higher than 10 for all the tests results, as indicated in Table 4.2 and Table 4.7. The experimental set simulating desert environment using sand and soil revealed that the C/N ratios were equal to 14.93, 14.99, and 16.79 for Sets, 1, 2, and 3, respectively, which was in the same range of comparable ratio of 11.76 using clay soil in the work of He et al. (2008). In all of these latter sets, the ratio is obviously high and could have been the reason for triggering the formation of EPS. The same high C/N ratios are also evident from Table 4.7, showing the results from the experimental sets 1, 2, and 3, investigating oxygen penetration. The ratio in the test beds of these latter sets of experiments showed an average C/N of 18.95, in comparison with Hilger's study, where it showed the average ratio of 13–22 (depending on the horizon level of the columns). Therefore, it could be concluded that these high ratios of C/N could have caused the formation of EPS in all of these experiments, indicating a stressing effect on the bacteria due to nitrogen limitation, and could have started the observed decline in efficiencies.

Material Type	Optimum MC (% wt/wt)	References
Landfill cover soil	15.6–18.8	Boecks et al., 1996
Landfill cover soil	18–24	Scheutz and Kjeldsen, 2004
Landfill cover soil	15–20	Czepiel et al., 1996
Soil	20–35	Bender and Conrad, 1995
Compost	21–28	Einola et al., 2007
Sandy Loam	15%+/- 5%	Hilger, et al., 2000
Compost and sand	29.60–31.46	Sets 1, 2, 3

Table 4.9: Materials and optimum MC compared with MC of the experimental Sets 1, 2, and 3, and Hilger's.

4.6 Type I, Type II, and Type X methanotrophic bacteria, their presence, and optimal conditions in column tests

In all of the aforementioned experiments, the ubiquitous methanotrophic bacteria were not identified. Each sample of these experiments contains multitudes of bacterial communities, living side by side. Therefore, to identify and categorise them in each of these sets and at each condition would require substantial investment in time and equipment, which unfortunately are not available. Nonetheless, published literature is full of information on the identity and the existence of these bacterial communities.

The aerobic methanotrophs, categorised as a physiological group Types I and II belong to the Alfaproteobacteria and Gammaproteobacteria classes, which use pathways pMMO and sMMO, respectively (Scheutz et al., 2009a). These two groups are the main classes found in soils and are responsible for methane assimilation. Besides these classes, there is the other group Type X (Methylococcus Capsulatus) categorised as a player in assimilating methane; although, this group is only active when specific conditions are present. This latter group is similar in physiological nature to Type I. Recently, this type has been grouped as subgroup of Type I (referred to as Type I-b). This species uses the same assimilation pathway (pMMO) to convert methane as that of Type I, but varying sometimes, in that, it uses the other sMMO pathway in their conversion (Hanson and Hanson, 1996). In addition, Type X group is active at higher temperature than the other two species and has higher mole percent than Type I with a higher ratio of enzyme pMMO over the sMMO (Knief, 2015; Lee, et al., 2015). Both species are found within the gamma subdivision. Type II, as previously noted in Section 2.2, employs the serine pathway for methane assimilation and coexisting as a coherent cluster within the alpha subdivision, as indicated in the genera classification of Table 2.1 (Hanson and Hanson, 1996, Scheutz et al., 2009a). This classification, however, is not a complete list of all the classes of methane-assimilating bacteria. More than 136 methane-assimilating bacteria have been identified in 1996 (Scheutz et al., 2009a), and more are being added continuously (Dianou and Adachi, 1999, Mehrabad, et al., 2012, Kalyuzhnaya, et al, 2016).

All of these species compete for their own energies and creation of their own biomass, utilising primarily whatever available methane and oxygen and whatever available organic and inorganic compounds in the soil. This competition is guided by their own physiological and genetic makeup, determining their own optimal operating limits and conditions accordingly. The guiding optimum environment for these bacteria to mitigate methane could be seen from published literature and are summarized in Table 4.10, which shows the distinctive difference between these groups.

Attributes	**Type I** (pMMO)	**Type II** (sMMO)	**Type X** (pMMO, sMMO)
Temperature	Low <10°C	High >20°C	High
Oxygen concentration (v/v)	High 21%	Low 1%	Similar to type I
CH_4 concentration	Low	High	Similar to type I
Copper (Cu)	High 1–5 µ mol/L	Low < 1 µ mol/L	Similar to type I
Nitrogen (Ni)	high	Low	Similar to type I

Table 4.10: Optimum conditions for the methanotrophic bacteria to assimilate CH_4 (Sources: Hanson and Hanson, 1996; Dunfield, et. al; 2007; Scheut, et al., 2009; Pfluger et al., 2011)

Methanotrophic bacteria Type I are active at optimum conditions of high oxygen concentrations and low level of methane; while Type II bacteria operate with optimum assimilation at low oxygen and high methane concentrations. These prevailing conditions would infer that a high rate of distribution of the Type I bacteria would be present at the top level of the soil, where oxygen is abundant and where methane has

low presence in the soil. On the other hand, Type II would be more active at the lower levels of low oxygen and high methane conditions. However, the optimum conditions shown in the Table 4.10 are not very decisive. Each type of bacteria could still be present in the distribution at lower or higher conditions than those shown, but they would assimilate at lower or higher efficiencies in a gradual process. For example, at temperatures lower than 20°C, Type II group could still convert methane, but with lower efficiency than they would if the temperature were at their optimum level, and *vice versa* for Type I. The same holds for each optimum condition specified, and for every bacterial species. Research would suggest that the most dominant factor in deciding the bacterial distribution along the depth of the soil is the oxygen concentration (Lee et al., 2015). Figure 4.21 shows an oxygen distribution pattern of a core sample of a rice paddy soil and a distribution of methane concentration for two soil sections, the rhizosphere soil (the narrow band of soil closely near plant roots), and the bulk soil (soil away from rhizosphere region). The rhizosphere zone provides organic carbon from the roots of the plants to the bacteria more than the bulk zones can do. The bulk distribution profile is considered to be the norm for oxygen and methane concentrations in the bio-cover soils of a landfill (high oxygen concentration at the top level and higher methane concentrations at the lower level (Lee, et al., 2015; Scheutz, et al., 2009b; Christophersen et al., 2001; Kightley et al., 1995), but with some variations in concentrations and depth, depending on the characteristics of the soil and the material used for the bio-cover.

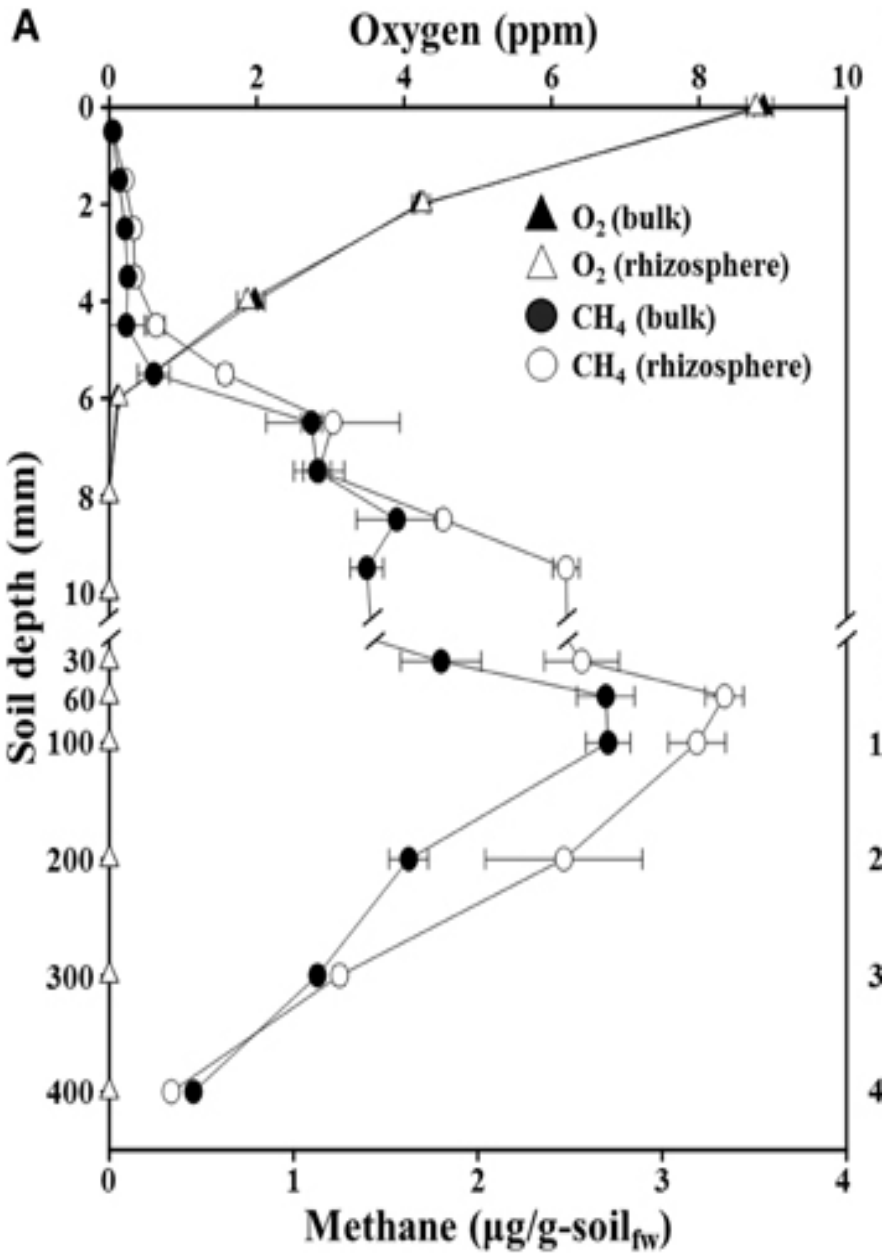


Figure 4.21: Oxygen and methane concentration of a core sample of paddy rice (source: Lee et al., 2015)

The patterns exhibited by the rice paddy samples would have a direct influence on the relative abundance of the bacterial cluster along the same height of the soil in a direct relationship (Figure 4.21). It is worth noting that Figure 4.22 shows this kind of cluster

distribution relationships, where seemingly, the methanotrophic bacteria have clustered along the same distribution line as that of the methane and oxygen distributions (Lee at al., 2015). The figure also shows that the relative abundance of Type II is much higher than that of Type I, outnumbering Type I all along the 400-mm core length of the soil (solid line in Figure 4.22 (A) and (B) differentiating the boundary between Type I, left and Type II, right). This observation is in line with the outcome arrived at by several researchers (Lee et al., 2015; Pfluger et al., 2011; Hanson and Hanson, 1996; Graham et al., 1993). The reason for this abundance is that Type II uses enzyme pathway sMMO, which has a broader substrate specificity than enzyme pMMO, and that Type II can assimilate methane at low oxygen and high methane concentrations than Type I can assimilate, in addition to the ability to outcompete Type I in an environment with limited nitrogen (Pfluger et al., 2011). Type X is labeled as a subgroup of Type I (Type I-b, light orange colour), abundant at the top of the soil sample, but decreasing rapidly with the depth of the soil. The figure also exhibits the abundance of methanogens, anaerobic unicellular bacteria, which produce methane from digesting organic compounds, existing all along the full height of the samples and even at deeper levels. These methanogens showed low relative abundance at the top of the soil but increasing substantially at the bottom, outnumbering the methanotrophic bacteria. This is however expected, since these methanogens are responsible for digesting organic matters, producing methane, and have low activity in presence of oxygen. The analysis of the soil samples in this example did not show any further bacterial groups existing alongside with this methane-bound species; nonetheless, abundance of other bacteria groups may as well exist in the soil, indicating the complex nature of the environment persisting in the soil.

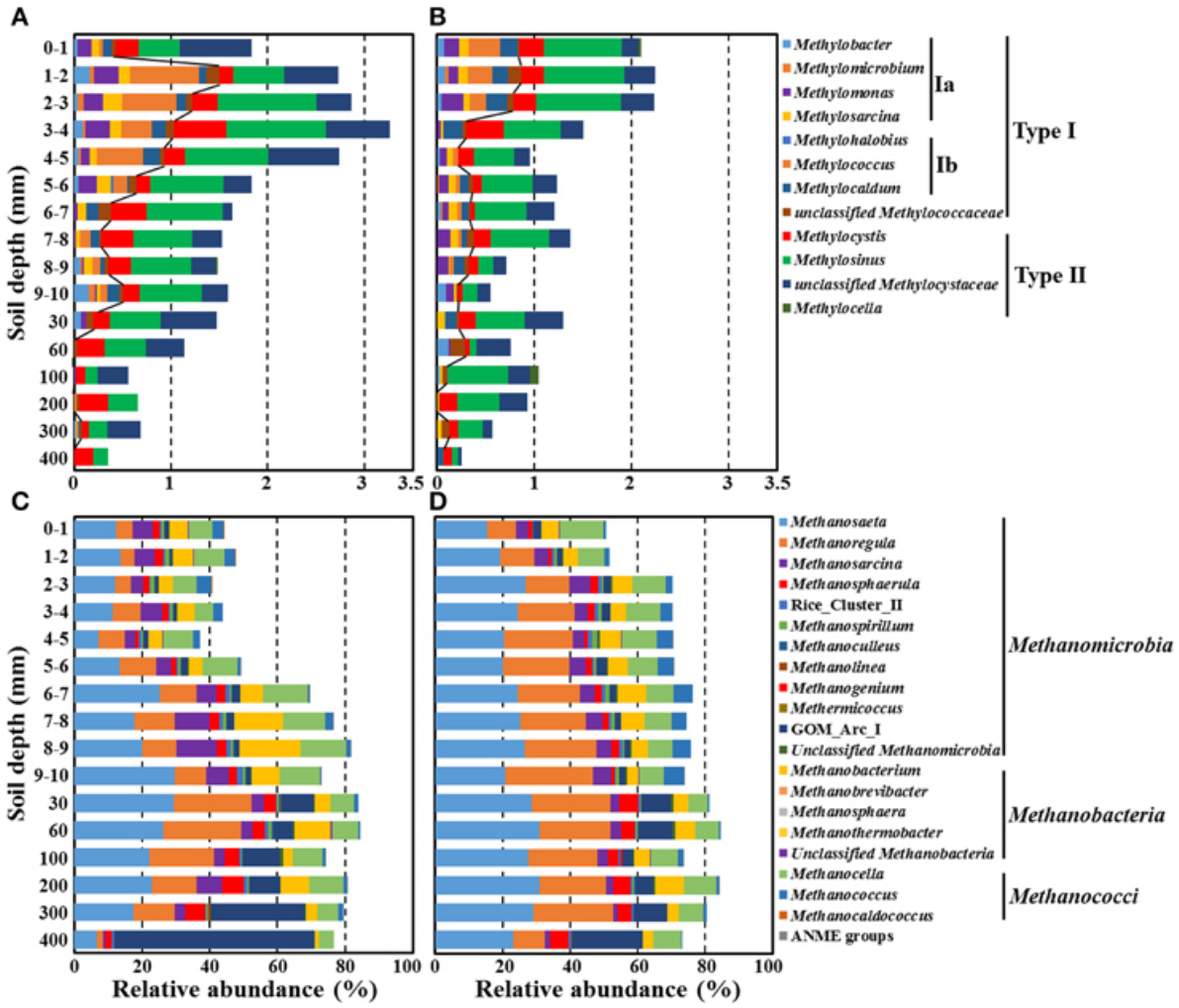


Figure 4.22: Relative abundance of methanotrophs A,B, and C,D for methanogens; for A,C bulk soil, and B,D rhizosphere soil (solid line in sub-figures A, and B differentiate the boundary between Type I, left and Type II, right; source: Lee, et al., 2015).

Based on results indicated in Figure 4.22, Table 4.10, the following can be concluded:

- A natural coexistence was seen to persist between the methanotrophic Type I (including Type X) and Type II along the depth of the samples in the column tests.
- A high relative abundance of Type II over Type I exists, due to their physiological cellular makeup, using enzyme pathway sMMO which has broader substrate specificity than enzyme pMMO.
- High probability of methanotrophs Type II to outcompete Type I is noted, when methane is limited.

- The extent of relative abundance of Type II along the depth of the soil is higher than that of Type I.
- Methanogens bacterial groups are present all along the depth of the soils, but mostly at the lower levels, where organic compounds are abounding, particularly if the organic compounds are supplied by the roots of the plants (rhizosphere soil region).
- The presence of methanogens at the high level of the soil is noticeable in the region where oxidation occurs, and much more than that at the lower end of the soil (bulk soil region).

The above conditions could have existed along the depth of each column of the experimental sets 1, 2, and 3, of each experimental trial discussed in sections 4.3, and 4.4, and could have influence the respective results.

4.7 Effect of organic amendments on oxidation

Data from published literature were extracted and compared against the oxygen concentration profile in the soil in order to show the importance of amendments to improving the soil's capacity to oxidise methane when organic matters (OM) were added to cover soils. Material containing OM in the form of coconut husk with an OM of 91.33% was added to a clay soil, characterised as 73.18% clay, 14.43% sand, and 9.9% silt. The oxidation results of a column experiment, using this type of material, were presented by Zainal and Buyong (2015) (Figure 4.23). Each curve in the figure represents a different ratio of OM to the soil, starting with samples of 0, 70/30, 50/50, and 30/70 of soil to OM ratios. In addition, a typical oxidation concentration profile of oxygen in soils is drawn along with the same plots to indicate the interdependent relationship between oxygen concentration and OM for methane oxidation (Humer and Lechner, 2001; Streese and Stegmann, 2003; Mostafid et al., 2012; Nguyen, et al., 2013). It was determined from these published literature studies that oxygen penetration on the surface of the soil can only be confined in the upper 30-50 cm, 15-40 cm, 40-60 cm, 15-40 cm, 3-12 cm of cover soil, as suggested by Visvanathan, et al. (1999), Nozhevnikova, et al., (1993), Barratt, (1995), and Whalen et al., (1990), respectively (Section E.10, Appendix. E).

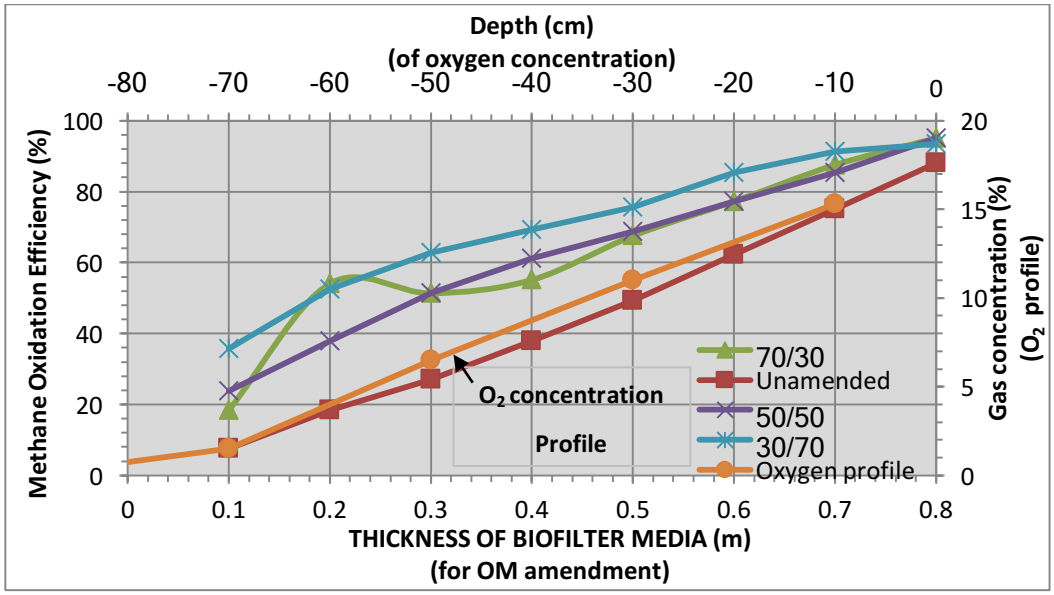


Figure 4.23: Oxygen concentration and oxidation efficiencies of CH_4 with OM additives, along the depth of a cover (Zainal and Buyong, 2015; Humer and Lechner, 2001).

Noticeably from the curves, the addition of OM to the soil in different ratios had a measurable influence on the efficiency of methane oxidation. This influence could be seen in all of the recorded data, even at a level of 10 cm from the bottom of the soil, indicating that even a small addition of OM in ratios of as much as 70:30 soil to OM, could have a positive effect. In contrast, the unamended soil sample produced only a 5% oxidation efficiency at that level; while the 50:50 and 30:70 samples resulted in an increase of oxidation efficiencies to approximately 21 and 35.5%, respectively, at the same level. At the top of the soil cover, the oxidation efficiencies of the samples showed good oxidation profiles, reaching up 97.5, 95.4, and 94.6% for samples 30:70, 50:50, and 70:30, respectively, with a steady decline moving from the top to the bottom. The curves of these amendments revealed almost the same trend as the trend of the oxygen concentration curve in the soil, indicating an overall interdependent relationship. These results are in line with the results obtained by Visvanathan et al. (1999) and Philopoulos et al. (2009), in their experiment using sandy clay loam and sand-perlite-compost materials, respectively. This led Gebert et al. (2011) to argue that the aeration along the depth of the soil, provided by OM amendment was the reason

for better oxidation efficiencies. Zainal and Buyong (2015) also suggested that the addition of OM had caused the increase in soil porosity and a decline in the bulk density of the soil samples from the unamended 1.3717 g/cm^3 to 0.3225, 0.4977, and 0.6814 g/cm^3 for 30:70, 50:50, and 70:30 soil to OM ratios, respectively, leading to high porosities and increased pore ratios.

The addition of OM to cover soils is important, particularly for dust-covered landfill cover layers, as the presence of OM permits easy methane transport from the bottom of the oxidation layer to meet with the easily diffusing oxygen through the pores created in the soil, thus, providing the methanotrophic bacteria with their needed share of oxygen and methane. The creation of these pore spaces is accomplished when the inner cells of the body of the OM decay. This is because the methanogenic bacterial action or chemical reactions leave the outer structure of the OM intact as support for those pore spaces to hold oxygen and methane. The OM mass content, in itself, may not be the cause of better methane oxidation observed in these experiments. Given that, at first, it could by itself produce methane under anaerobic conditions due to the decaying biological matters of the biomass, if not weathered enough; subsequently, whenever the state of the OM materials reaches maturity, the materials become useful. Therefore, the oxygen delivery venues created by these structures allowing the movement of gases to and throughout the oxidation layer are deemed responsible for the increase in the oxidation efficiencies of methane. In addition, OM addition could provide needed biological nitrogen compounds to the soil for the use of the methanotrophic bacteria. The experimental tests discussed in Section 4.2, where the OM levels were 18.56, 3.62, and 0.67% (wt/wt), evidently, have indicated that these low levels of OM in comparison to 91.33% (wt/wt) OM amendment to soil reported by Zainal and Buyong (2015), did not have positive effects on methane oxidation.

4.8 A conceptual design of a system for the mitigation of methane in arid environment

Based on the results gained from the continuous flow experiments, Sections 4.2 and 4.3 and from the information gained from reviewing the literature, it appeared that methanotrophic bacteria cannot function properly if oxygen diffusion was somehow hindered in the soil, due to regulatory or environmental conditions. Therefore, a new alternative system must henceforth be devised to allow better oxygen diffusion. This alternative system consists of funnel/pipe, with a regulating gate system designed to provide the essential air inside the oxidation layers for the bacteria, without disturbing the regulated and avoiding the blocked top covers. The makeup of this proposed system is shown in Figure 4.24 and described in detail in Appendix D. The system allows air to be delivered inside the oxidation layer by the action of wind force using a funnel. The funnel size is dependent on the site wind and geographical attributes, which pass air through a one-way gate system and through an immersed pipe in the soil to deliver the needed oxygen to the methanotrophic bacteria.

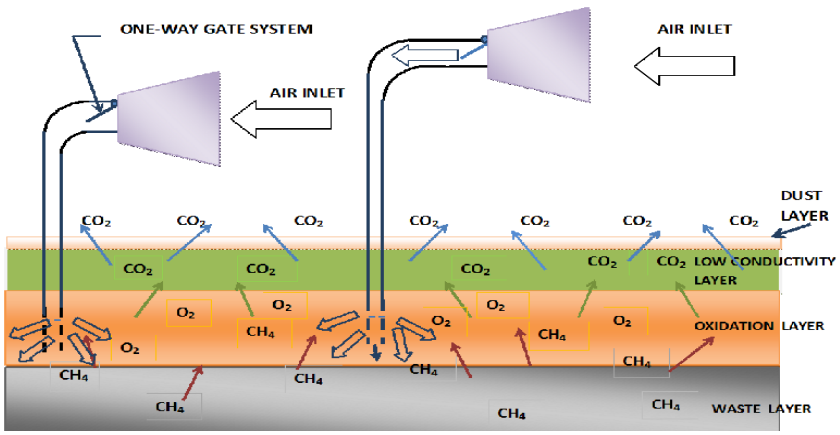


Figure 4.24: Conceptual design of a funnel system for mitigating methane.

Meanwhile, in order to show the applicability and effectiveness of this system, a numerical simulation model must therefore be done. The simulation of this system would not be rendered useful unless the outcome of that simulation is compared to actual field data.

4.8.1 *Field data of the Kelso Waste Depot Landfill, Sydney, Australia*

Field trial data for an experimental bio-window system, constructed on Kelso Waste Depot Landfill in Western Sydney, Australia, with an extensive trial that lasted for 6 years (Dever et al., 2006, 2011, Dever, 2009), were used to compare with the simulation model. Data for this bio-window filter used for this field experiment consisted of four 1.2 m in depth and 3 m by 3 m wide filter cells, constructed alongside each other with each bed of filters, made of different materials. The data for one cell (a bio-filter D which was made of composted solid waste with 10% of shredded wood) showed that the landfill gas was flowing at a rate of 0.26–1.0 m^3h^{-1} and composed of methane and carbon dioxide at a ratio of 61.6% to 34.3%, respectively. The data also showed that the soil was of clay/shale characteristic, having hydraulic conductivity of approximately 1.75 $x10^{-5}$ m s^{-1}, and possessing a porosity of 49.5%. The prevailing atmospheric conditions at that site were variable, measuring at an average atmospheric pressure of 101.7 kPa and of a variable surface wind speeds at a range of 0 to 3 km/h.

4.8.2 ***Numerical simulation of the system***

Using the data of the forgoing section, a cell of 3-m width by 1.2-m depth was created, to simulate the cover of the Kelso Waste landfill (a bio-filter cell D containing composted municipal solid waste amended with 10% shredded wood), using advective methane/carbon dioxide gas input with an average composition ratio of 61.6/34.3, flowing with a high end rate of 1.0 m^3h^{-1}. The schematic diagram of the model is shown in Figure 4.25. This gas flow rate was the maximum rate recorded for that filter in that field landfill experiment. The simulated boundary conditions of the cell were 101.7 kPa, as applied atmospheric pressure on the top surface and a wind blowing at the speed of 0–3 km/h through the funnel, producing pressure at the end tip of the pipe in accordance with Bernoulli principle (Appendix D). These boundary conditions data are the same data present at Kelso landfill test ground and indicated in Figure 4.25. Funnel gate mechanisms was not modelled in this simulation, as well as the bacterial kinetics, due to the inability of the numerical code to mix the mechanical

dynamics and fluid flow functions together, or simulate single cell biological entities. However, the wind pressure can easily be calculated and entered as a variable input inside of the pipe, simulating the functions of the gate and the funnel system. To simulate this cell, a finite element program code was used.

The finite numerical method is a powerful and well established technique developed originally to analyse structural systems, and has been extended later to solve fluid and heat transfer problems. This numerical technique, in simple terms, relies on the process of dividing any structural (or fluidic) systems to very small microelements, having the boundaries of each element to continually match the boundaries of the other surrounding elements (by way of the continuity equations). When these elements are numerically added, they would congregate to simulate the whole body of the complete structural system, and then, the complete analysis would emerge when the external forces and external boundary conditions are added. Thereafter the accuracy of such representation depends on the number of elements, with the divisions, being dictated by the level of accuracy required. However, that would be on the account of cost, time, and computer storage space. Another important advantage of this numerical technique, besides the level of accuracy, is being able to represent any system of any shape or form by having fine subdivisions of that system to a fairly accurate representation (Reddy, 1993, Zienkiewicz, et al., 2006). This same accuracy of representation is transferred subsequently to accuracy of the results outcomes. This representation is used to simulate the Kelso biofilter trial, for the immersed pipe, as well as for the body of the filter of Figure 4.25, using the powerful pre-processing mesh generating utility of finite element program code ANSYS. For modelling this biofilter, the pipe element of the system was simulated as a conduit having a width equal to the diameter of the pipe, simulated as a requirement of the 2-D modelling. For the 3-D modelling, however, the pipe element would then be modelled as a pipe element, with radial outflow in 3-D space.

One of the basic requirements of the theoretical development of the finite element technique is to keep the finite elements intact to preserve the integrity of the system. This would mean that the elements are attached via nodal elements to one another, to

keep in line with the mathematical rules of continuity. For systems having loose, unattached, consuming, and producing their own materials, dividing, living, and dying, and in essence, having their own physical and physiological rules, such as bacterial communities existing in the soil, these conditions cannot be represented by the same mathematical rules bounding structural systems (Zienkiewicz, et al., 2006). Therefore, the kinetics and activities of bacterial community, residing in the Kelso biofilter system cannot be simulated or modelled by any finite element code, including the very developed ANSYS code, as the simulation of such biological behaviour is not available (ANSYS Verification Fluid Dynamics Manual, 2011). Nevertheless, the activities of such bacterial community are dependent on the oxygen and methane concentrations, as shown by Cao and Staszewska, (2011) (Figures E.7, and E.9, Appendix E). Consequently, their level of activities in the soil can be predicted by these graphs.

This powerful analytical finite element program employs a user-friendly interface, allowing users to enter data via clearly explainable windows. The modelling starts with a pre-processing interface that aids in creating finite elements needed for modelling objects, and then, transfers the results to another processing utility that can analyse the object's attributes, and solve for required output parameters. For modelling the Kelso biofilter, the basic data, as feed data to the pre-processing utility of the program, such as the dimensions, material type, boundary constraints, properties of the porous medium, fluid properties, external velocities and pressures, are all given in to the program. The pipe well in this simulation was modelled as a conduit of two walls, placed inside the porous medium, having impermeable boundary, and a width equal to the diameter dimension of the pipe (similar to the simulation of the two boundaries of the soil cell). This two-dimensional treatment of the pipe system allows placing flexible pressure force on the air inside the top boundary of the conduit, and allowing only two-dimensional fluid flow inside the porous medium. Once all the data are entered, the post-processing utility of the program brings out the results in velocities, volumes, forces, displacements, and can graph the results, as well. The code manual is very comprehensive, aided by multitudes of tutorials, and supplemented by a rich resource of internet video examples (ANSYS Verification Fluid Dynamics Manual, 2011).

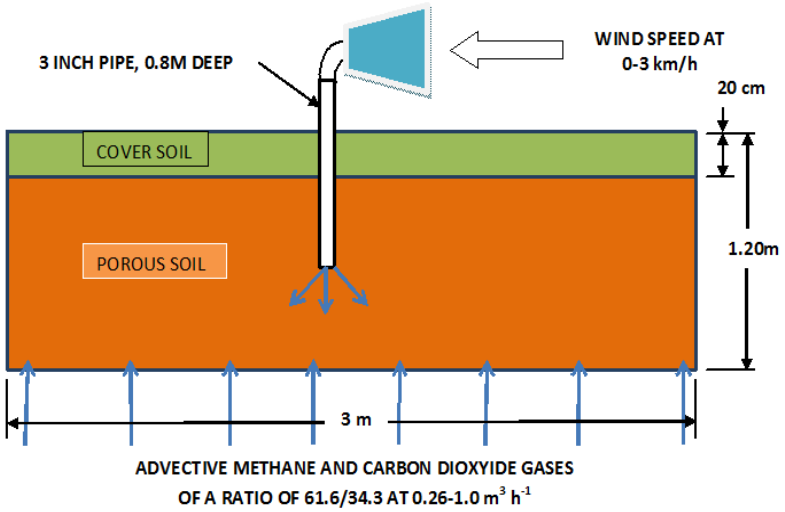
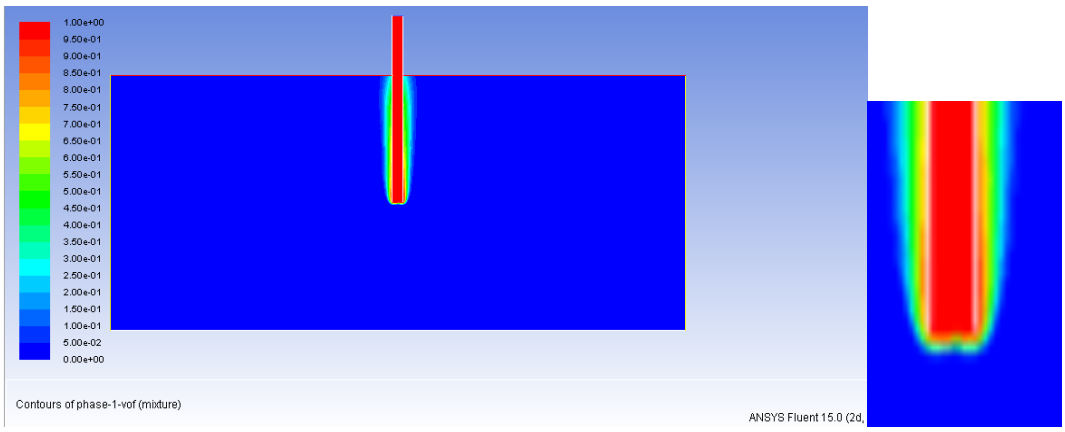


Figure 4.25: Diagram of the simulated model.

In this model, when the wind speed was kept low (i.e. 0), no additional pressure at the tip of the pipe was produced beside the atmospheric pressure. The simulated model showed that methane gas was flowing throughout the porous medium of the cell, although faster in the pipe venue than in the soil medium itself. This was observed in the finite element program produced model in Figure 4.26. It is imperative therefore, that the pressure inside the pipe be increased in order to overpower the pressure of the advective mix of gases inside the porous cell medium. This increase in pressure would require an appropriate funnel size to be used for this higher pressure. For a speed of 3 km/h above the surface of the landfill, the funnel size diameter to the pipe diameter (D_1/D_2), needed to just balance out the pressure inside the cell, was estimated to be 11.22 (using Bernoulli principle, Appendix D, equation D.23). Figure 4.27 shows this case of just allowing enough air pressure inside the pipe if only to overpower the internal gas pressure in the porous medium. However, when the funnel to pipe diameters was increased to 17.72 and 25.89, the pressure inside the pipe increased, consequently, allowing substantial amount of air to diffuse inside the porous soil medium (Figures 4.28 and 4.29, respectively). These figures simulated air diffusion in a steady continuous flow state; while advective methane and carbon dioxide gases diffused inside the medium also in steady flow state. The pressures induced landfill gas

and air throughout the soil medium when the funnel to pipe ratio was at D_1/D_2=25.89 is shown in Figure 4.30.

Figure 4.26: Molar concentration of CH_4 in the porous cell medium, subjected to only an atmospheric pressure (D_1/D_2=0), with the gas flowing up through the pipe. (Scale in units of K mol/m^3)



(a) (b)

Figure 4.27: Volumetric air diffusion (% v/v) inside porous soil medium of a funnel to pipe ratio of D_1/D_2=11.22, for a flow of just overpowering the internal gas pressure (a) full length profile, (b) pipe end.

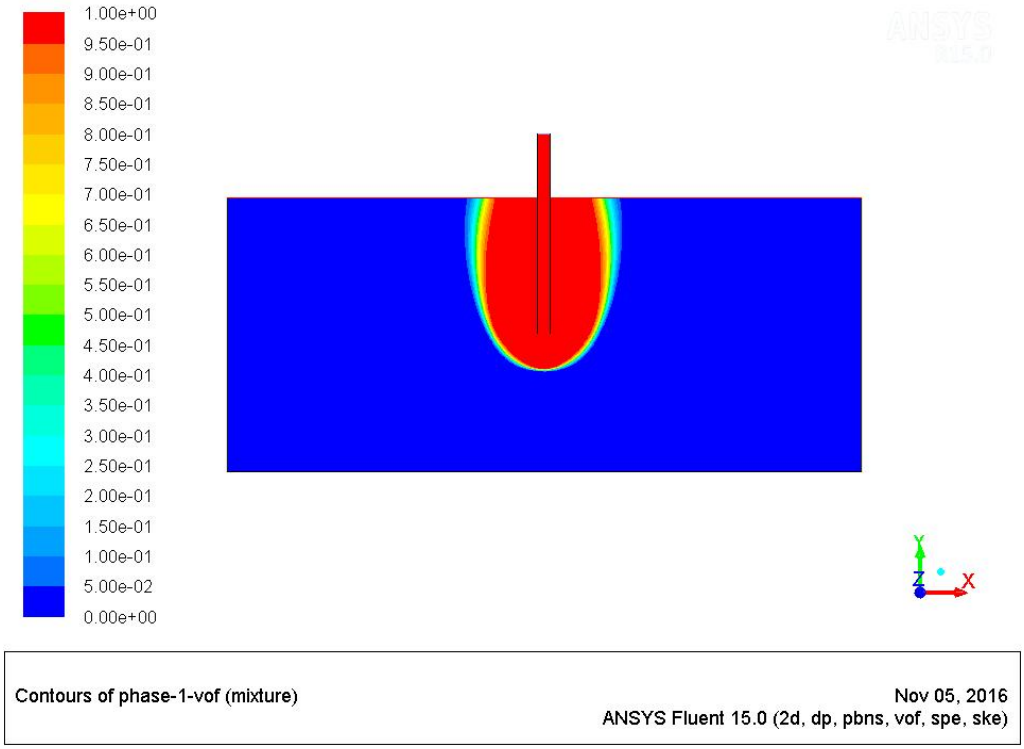


Figure 4.28: Volumetric air diffusion (% v/v) inside porous soil medium of a funnel to pipe ratio of D_1/D_2= 17.72, with a high advective LFG of 1.0 m^3 h^{-1}, in simulation to Kelso Waste landfill.

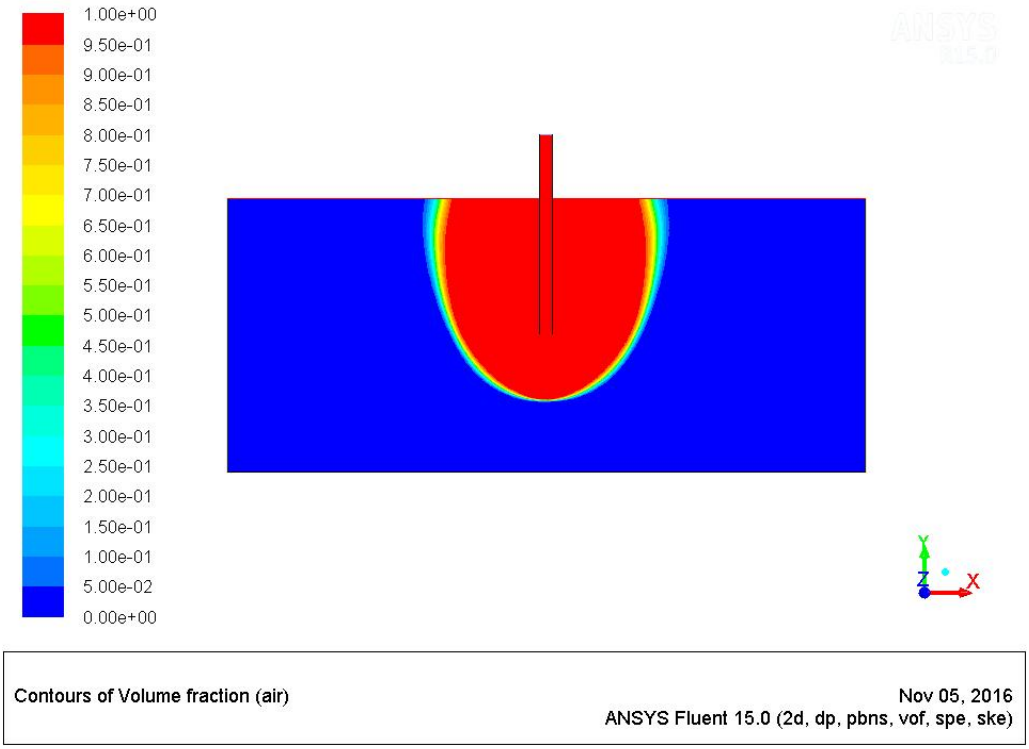


Figure 4.29: Volumetric air diffusion (% v/v) inside porous soil medium of a funnel to pipe ratio of D_1/D_2=25.89, with a high advective LFG of 1.0 m^3 h^{-1}, in simulation to Kelso Waste landfill.

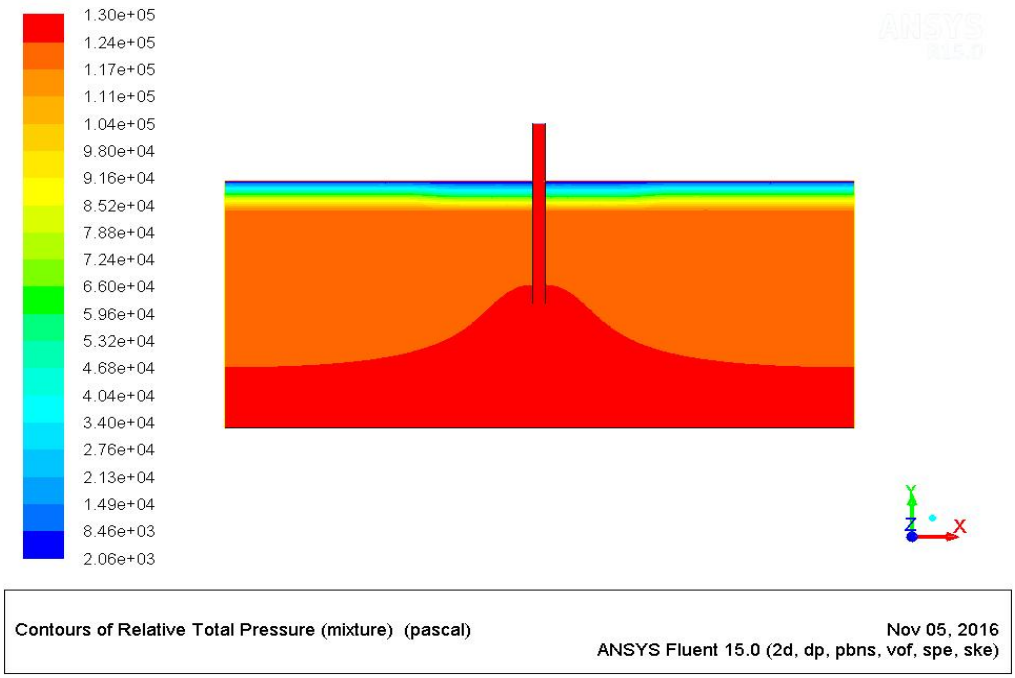


Figure 4.30: relative pressure inside the porous soil medium subject to the atmospheric pressure and to the pressure due to funnel to pipe ratio of $D1/D_2$=25.89. (Scale in units of Pa)

4.8.3 *Comparison with Kelso Waste Landfill trial field data*

It was observed from Figures 4.26, 4.27, and 4.28 that when the pressure in the pipe was increased, due to a high funnel to diameter ratios, the flow of air had increased inside the soil medium, accordingly and around the pipe in an upward movement, rather than in a downward flow. This however is expected, since the moving advective methane and carbon dioxide gases carried along in their path the air molecules in an upward motion. When the pressure inside the pipe was sequentially made higher from a ratio of D_1/D_2 11.22 to a ratio of 25.89, the higher ratio allowed deeper air penetration to reach a distance from the surface inside the soil from 0.6 m to 0.91 m deep (Table 4.11), and also widened the horizontal cross-sectional area from a diameter of 0.95 m to 1.46 m, respectively. By way of comparison, Kelso's field data for a bio-window filter, made of composted MSW with 10% shredded wood, showed that the maximum oxygen penetration could only be found at few centimetres deep from the surface of the soil medium and reaching a maximum penetration of 0.2 m deep (Figure 4.31). The addition of shredded wood in this field filter trial was meant to

increase the void spaces in the filter's bed, and therefore, increasing the amount of air penetration inside the filter, as this is the standard practice used in these new filtration systems. The Kelso data can be compared with the diffusion of oxygen inside the simulated model, where the penetration of two simulated cases had shown a substantial difference (Figure 4.31 and Table 4.11), where higher funnel to pipe diameter ratios produced deeper and wider oxygen availability to the bacterial community in the soil than would a bio-window system. Air penetration curves of the model shown in Figure 4.31 were taken from across the depth of the model and exactly adjacent to the pipe. ANSYS program code can bring out the measurement of air penetration in %v/v of air at any cross section. From these measurements, air ratio was at 100% (of which 21%was oxygen) at the top surface and continued with that same ratio down across the section, until suddenly reduced at the boundary of contact by the upflowing mixture of methane and carbon dioxide gases, as clearly shown in Figures 4.28 and 4.9. This air profile across the depth came out to be in that form as a result of the steady state balance between air diffused down from the top, and the mixture of LFG gases moving upward. The level of penetration, reaching 0.91 m, compared to only a maximum of 0.2 m of the Kelso data would be more than four times the level of penetration over the bio-window system used in the field trial. However, the variables of the simulated model did not exactly represent the actual field variables, as the case of most simulation models. For example, the granular makeup of the soil, which was not generally homogenous in nature, was assumed to be uniformly homogenous throughout the soil medium, with all of the other dependent soil variables assumed to be constant across the medium.

Funnel to pipe diameter ratio (D_1/D_2)	Penetration Depth (M)	Width of penetration (m)	Wind speed V (km/h)	Advective gas flow ($m^3 h^{-1}$)
0	0	0	3.0	1.0
11.22	0.6	0.29		
17.72	0.74	0.97		
25.89	0.91	1.46		

Table 4.11: Maximum depth and width of air penetration for different funnel to pipe diameter ratios.

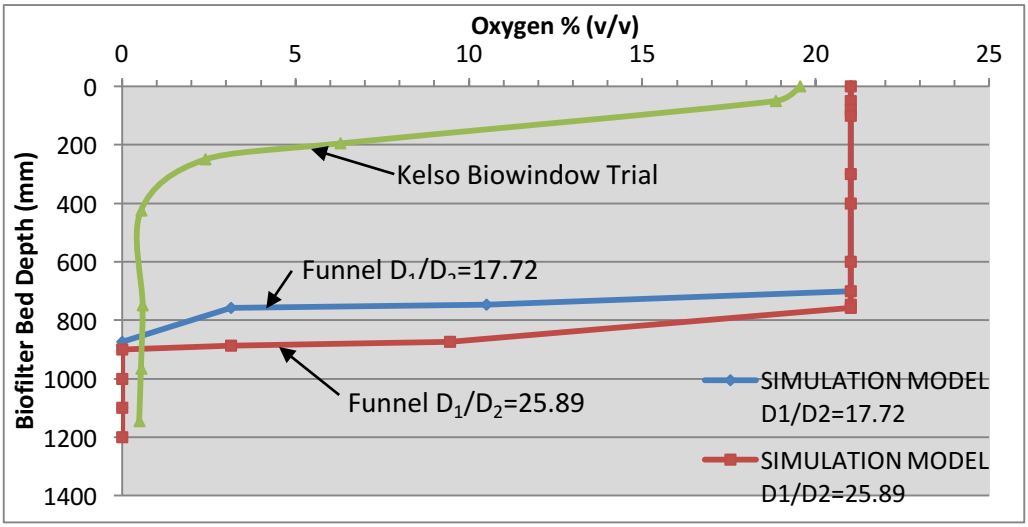


Figure 4.31: Oxygen penetration in bio-window filter of field data conducted on Kelso Waste Depot Landfill, Sydney, Australia (Dever et al., 2011), in comparison to the oxygen penetration levels data for a funnel simulation model.

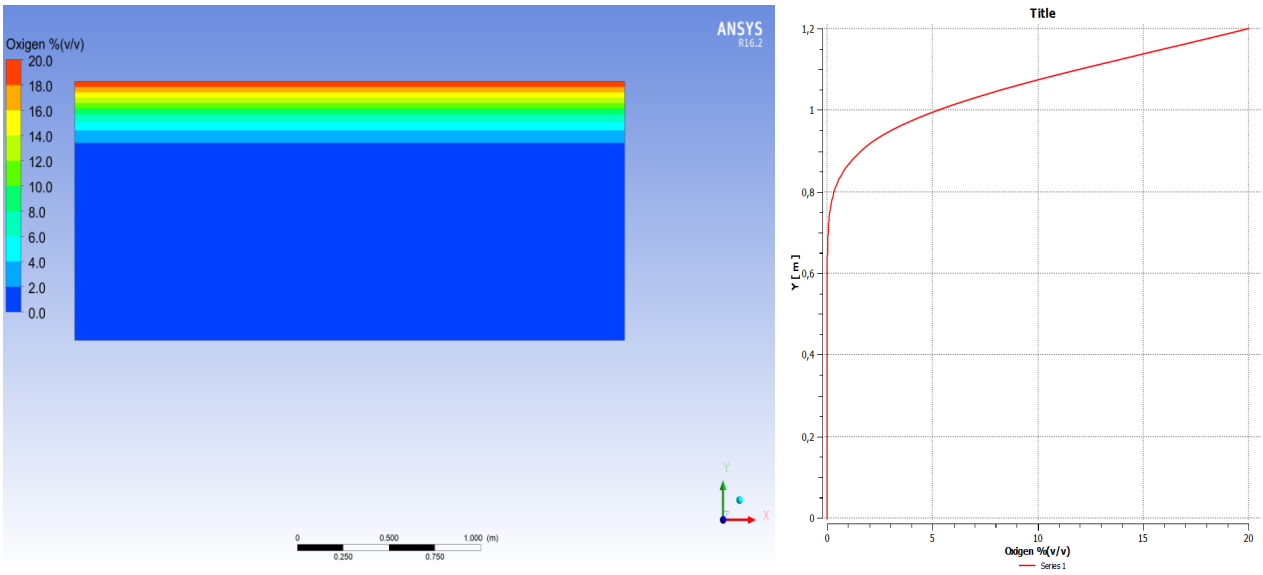
Data of the Kelso Waste landfill oxygen penetration shown in Figure 4.31 indicated that the oxygen distribution inside the filter was due to the action of atmospheric pressure fluctuations, moving air molecules up and down, causing gradual distribution of oxygen throughout the soil medium. On the other hand, the data for the simulated model showed only the static conditions of the two-phase flow gases in balance. Each curve of the simulated cases (Figure 4.31 and Figures 4.26–4.29) represents a balance of the pressure forces of air coming from the pipe, methane, and carbon dioxide diffusing from the bottom of the soil's cell in a static balance. However, in natural settings of landfills, this balance of pressure forces becomes dynamic, fluctuating in accordance with wind force fluctuations, pushing air molecules up and down the soil in a similar fashion to the atmospheric pressure action on the bio-window in Figure 4.31. Henceforth, the bio-window filter and the funnel systems have their own pumping mechanism of air into the soil. The wind speed fluctuation in the simulation model, causing fluctuations inside the soil medium, could range from the case of zero wind speed (Figure 4.26) to high wind speed (Figure 4.29). Therefore, curves of Figure 4.31 and the diagrams of Figures 4.26, 4.27, 4.28, and 4.9 represent the extent and bounds of oxygen distribution inside the soil medium as a result of varying funnel sizes in simulation to the Kelso field data. Providing substantial air into the soil affects the

number and activities of the methanotrophic bacteria in a direct way and according to Figure E.7 (Appendix E).

It is clear from the aforementioned analysis that there was a substantial penetration advantage of oxygen in favour of the funnel system over a bio-window filter; however, the comparison of two systems that are of entirely different designs may not be justifiable. Firstly, the physical-biological complex systems that are co-existing and interacting with each other in a landfill soil are difficult to simulate exactly using a simple numerical code. Secondly, the bio-window system depends on atmospheric and molar diffusion mechanisms, which are both unreliable and inefficient. Once a bio-window system were to be constructed over a soil medium, the variables of that system would not be flexible enough to be altered or changed to produce efficient performance. For example, the depth of the bio-window cannot be changed once it is constructed; the dimension and material type used in the filter bed are also difficult to change. Additionally, larger-sized bio-window system entails high cost of materials and high construction efforts. In contrast, the diffusion variables of the funnel system can be readily altered. Variables, such as the diameters of the pipe, diameter of the funnel, pipe insertion depths, number of funnels inserted, height of the funnels to catch higher upwind speeds, and direction of the funnels could readily be replaced, changed, or altered to suit the evolving environmental conditions persisting in the site. All of these changes could be achieved without altering the regulated or blocked surface of the landfill, thereby, increasing the efficiency of methane elimination with a high degree of flexibility. Moreover, addition of any number of funnels on the surface of landfills to cover the entire surface, for maximum penetration can be achieved with minimum investments in efforts, materials, and time. Therefore, these two systems are equally distinctive to warrant close comparison, and the results can only be taken as indicative of the performance of methane elimination of each system. A much more dedicated study to compare performance of each system on an active landfill, which could include experiments and simulations, would be an interesting future project.

To verify the results of the model against the actual results of the Kelso bio-window filter trial, the numerical model was again analysed using the ANSYS program code, this

time without the air injection pipe. To have the pipe well inside the soil domain, even without air injection, would provide an easy access for the gases to escape from the soil to the atmosphere. Therefore, it was deemed necessary to produce the same design setup as that of the system installed in the Kelso biofilter trial; hence, the results of both setups should reveal similar outcomes when compared. Figure 4.32 shows the outcome of the numerical simulation model of the biofilter, indicating oxygen distribution flow inside the biofilter bed, while Figure 4.33 shows the results of the diffusing mixture of methane and carbon dioxide gases throughout the bed medium. In both cases, the gases were in steady state condition. The distribution profiles of oxygen (O_2) and the gas mixture of methane and carbon dioxide (CH_4+CO_2) across the depth of the simulation, entering as a mixture gas in the model, are shown in Figure 4.34. This output was compared with the Kelso bio-window filter data (Dever, et. al, 2009, 2011), and the results of this comparison showed close agreement between the two setups. Therefore, this agreement validates the use of the numerical model in simulating Kelso landfill biofilter.



(a) **(b)**

Figure 4.32: (a) Oxygen distribution profile in the numerical model simulating Kelso biofiter (b) graphical representation of oxygen profile across the depth of the model %(v/v).

Even though the numerical model used in this study was just a theoretical representation, and was able to simulate the Kelso biofilter to an extent, nevertheless, validation of the injection model of Figure 4.25, to simulate the complex environmental conditions of landfills, still requires further experimental validation. This validation should be carried out on local landfill sites.

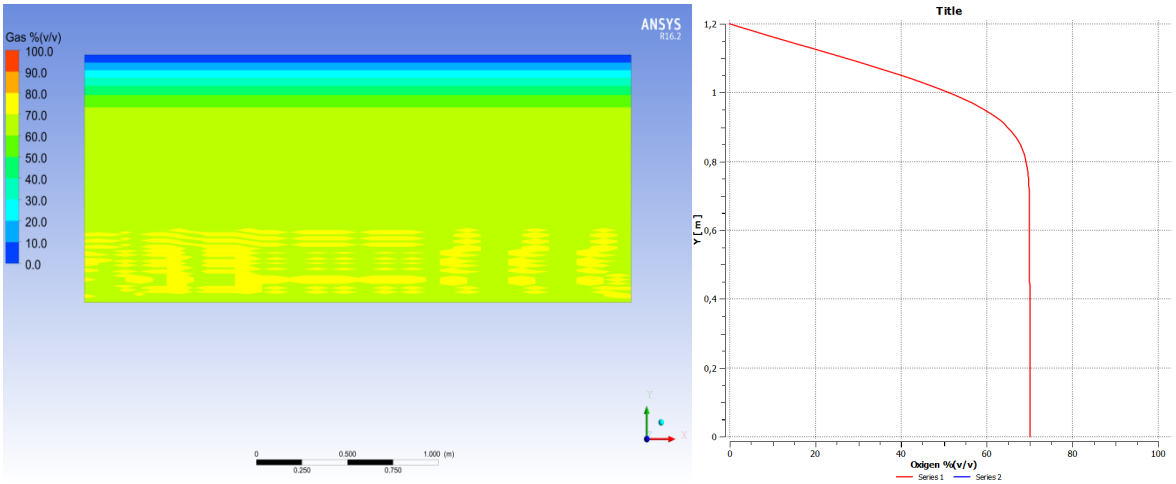


Figure 4.33: (a) Gas (CH_4 and CO_2) distribution profile inside the numerical model (b) graphical representation of gas mixture profile across the depth of the model %(v/v).

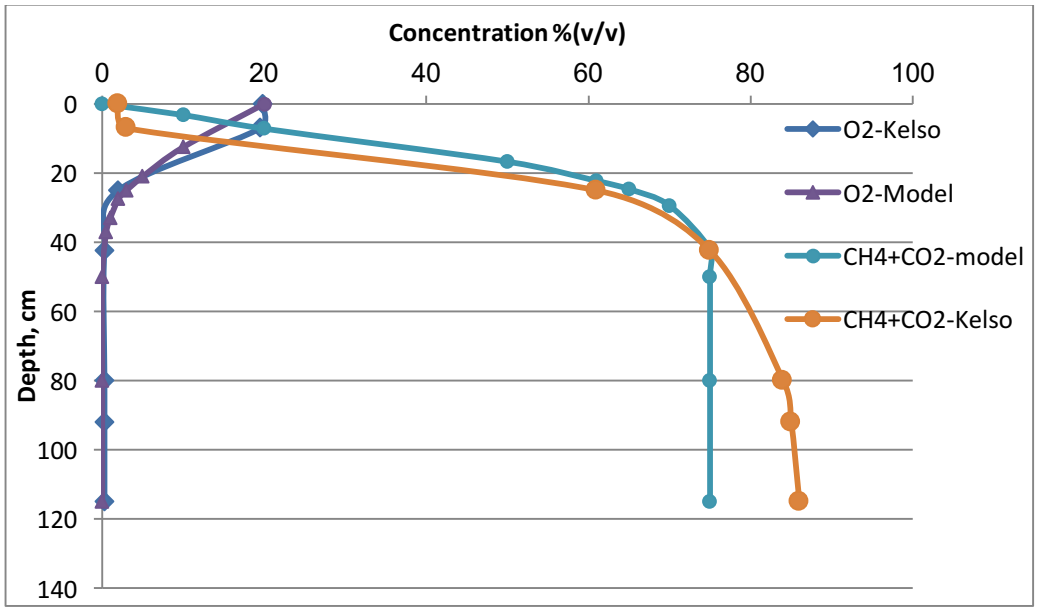


Figure 4.34: Comparison of the results between Kelso biofilter trial and the numerical simulation model of the filter.

Chapter V
Conclusion and Recommendations for Future Research

Constrained by the inability of conventional and improved landfill technologies to properly eliminate landfill methane and in response to the ever-increasing international pressures to clean up global carbon footprint, using the persuasion of taxations and the enactment of other various regulations, landfill operators in Kuwait are in need of understanding and finding alternative methane control methods to manage and restrain their particular environmental condition.

Findings in the literature showed that the major thrust of the research was mainly concentrated on enhancing air diffusion into the soil of the landfill cover, of which the bulk of it was dedicated to finding the most suitable cover materials to enhance the efficiency of oxidation, as indicated in Chapter II and Appendix A. Material aggregate sizes, types, orientations, maturity, composition, additives, compaction, porosity, conductivity, etc., were factors experimented extensively for the sole purpose of increasing efficiency by way of increasing voids in the soils and pockets inside the cover to encourage air penetration (Appendix E). Organic matter was also studied for the sole purpose of increasing the air pockets inside the soil, given that dead cells leave their organic structure intact when the biomass of the cells are consumed by bacterial or chemical reactions inside the soil, leaving their cellular structure intact to be filled with the diffusing air (Section 4.7). Similarly, moisture contents were also dealt with in the literature studies for the purpose of preventing water from occupying the very important void spaces existing between the grains of the soil, and hence, preventing air from diffusing to the inside of the soil. Among others, atmospheric, and wind turbulence were factors that affect air penetration directly; however, they have been proven to be inefficient (Sections E.3, E.6, E.10, E.21, Appendix E). Therefore, all of these studied factors were just means of delivery of oxygen to the inside of the soil, and given the particular environmental conditions existing in Kuwait, oxygen availability and delivery to the inside of the landfill soil was similarly investigated in this study (Sections 3.4.4, 4.3), however, in a more direct approach. Therefore, it can be concluded from

these literature studies that they were concentrating on satisfying the bacterial requirement of CH_4, O_2, and NH_4, according to the bacterial reactive equations (2.1 and 2.2), out of which O_2 becomes the most important factor for any bacterial activities to occur.

Moisture contents, temperature, nutritional organic matters, and diffusion of air inside landfill soils are major factors that affect air penetration into the soil, particularly in arid zone climates. Although rain precipitation in the desert climate of Kuwait does not exceed 100 mm a year, suggesting dry landfills' soils, some studies (Al-Yagout et al., 2007) have revealed that moisture contents were sufficient enough to warrant unhindered oxidation. Also, moisture contents accumulated in response to the high concentration of organic matters and high volumes of liquid deposits in these landfills of Kuwait were known to range between 15 and 40% (wt/wt), a concentration falling within the range of optimum methane elimination. The other methane controlling factors, such as pH levels and temperatures, were also measured at these landfills, showing pH levels to range between 7.82 and 8.00 and a temperature in the range of 23 to 35°C, even at high atmospheric temperatures, inferring that these factors are within the allowable limits for efficient methane oxidation in these landfills.

The third most important factor, besides moisture contents and temperatures in soils of arid environments, is the provision of organic matter inside the oxidation layer of the landfills. Amendments to landfill cover soils were intended, firstly, to enhance air penetration into the soil, and secondly, to provide nutrition in the form of organic ammonium (NH_4^+), and to provide other important nutritional elements, such as copper, potassium, iron, etc. For that purpose, the most used cover amendment was compost material due to its quality to provide better air diffusion and organic nutrients at the same time. However, compost material must be fully matured so that it does not produce its own methane gas through an anaerobic process. The evident conclusion relative to this maturing process, is that bacterial degradation of organic matter (or chemical interaction) would decompose organic cells to basic minerals, leaving only the outer shells of the cells intact, providing venues for air diffusion. The use of this diffusion characteristic of compost material, or of any other amendment materials with

the same quality for surface diffusion, is unattainable if used in arid environment due to the poor surface vegetation, and the unavailability of compost producing facilities. However, the provision of organic nutrients, as another characteristic of the compost material, is essential for the desert environment. Investigations and researches on nutrient additives, such as nitrates and ammonia are abundant. It has been reported that less than 25-g N (nitrogen-based compounds)/kg of soil could encourage oxidation; whereas, higher amounts could encourage high conversion rates of ammonium (NH^{+}_{4}) to NO_2, in a direct competition for oxygen with other bacterial communities present in the soil. Methanotrophic bacteria have high requirement for nitrogen, as they need one mole of nitrogen for every four moles of carbon up to certain concentrations. When the molar ratio of carbon to nitrogen (C/N) exceeds 10, nitrogen becomes limiting. Unfortunately, this ratio level of carbon to nitrogen is not likely to occur in landfill in desert environment, due to the poor vegetation available on the cover. Even if direct air (having nitrogen) is allowed through this impermeable top cover layer, where methanotrophic bacteria type II can assimilate nitrogen from air, it is more energy-efficient for the bacteria to use the reduced nitrogen available in the soil. In the process of using nitrogen compound nutrient, methanotrophic bacteria (type II) can assimilate nitrogen from the inorganic nutrients either in forms of nitrate or in forms of organic ammonium, using their particular monooxygenases (pMMO) enzymes for nitrogen fixation. Even so, the availability of organic matter is important to improve the impoverished arid soils, so is the other factors, such as air diffusion to the inside of cover layers, and the type of soils of desert nature covering the wastes. All these factors need further investigation. Therefore, batch and column tests were conducted to measure the effects of these factors, namely, oxygen availability, continuity, and low organic matter contents in desert soil on methane oxidation.

The first objective of this study was to examine the behaviour of methane oxidising bacteria subjected to conditions of oxygen availability and sustainability. The batch experiments carried out in this study showed that oxygen was an essential element in catalysing methane through the methanotrophic process. The bacteria metabolised methane through the breakup of its molecules in the presence of oxygen, therefore, generating their own energy and their own body biomass in a well-known process. The

most important element in this process, however, was the sustainability of oxygen in the soil within a sufficient period to catalyse the methane gas. These conducted batch experiments have shown a dramatic increase in bacterial consumption of methane when the soils were placed on top of shaking platforms, allowing oxygen to penetrate the soil grains deeper inside the soil reaching further bacterial communities. Thus, by providing oxygen and nutrients to these bacterial communities, an increase in the consumption of methane more than twice as much as the consumption of the samples placed on unshaking platforms was observed (Sections 3.3.4, 4.1.3). In addition, when oxygen was injected in the batch chambers within two separate time intervals, the consumption of methane continued linearly with time, suggesting that methane could be completely consumed if oxygen were to be introduced continuously. This is indicative of the importance of sustainability to provide oxygen into the catalysing cover soils of landfills. Additionally, the experiments showed that almost all soil samples, regardless of their composition or their degree of exposures to the methane gas, reacted in a very similar way under the oxygen availability. This is to infer that oxygen should be the primary and dominant factor to be considered when designing a bio-cover system for landfills in arid environment.

The batch reactor experiments also showed that three to four days of time lag must elapse before an active oxidation can occur, particularly if the material chosen for a cover had little or no methane exposure. In this scenario, more methane gas would escape from the landfill before the methanotrophic bacteria could have the time to regenerate in that cover soil; meaning in that, more unchecked methane would escape into the environment. For landfills to operate for approximately 20 years before closure, according the US-EPA landfill model, substantial amounts of methane per day globally would escape, constituting an important element of carbon balance to be considered by landfill operators. To combat this time lag, a methane-exposed soil would be suitable for immediate installation instead of the common practice of using whatever soil material available on site to cover the wastes. However, this step had to be addressed carefully, since premature and highly exposed soils would produce their own methane through anaerobic action and could inhibit the methanotrophic bacteria, particularly when nitrogen compounds are present (Bodelier and Laanbroek, 2004); this

in turn would produce more methane into the environment. These two extreme conditions may be resolved by having an appropriate cover material, matured, and does not produce its own methane.

The second objective of this study was to investigate the condition of suitability of commonly available soil material in desert environment, as a cover layer, on methane reduction. Simulating the environment of desert's soil materials, soil and sand mixes were tested for oxidation efficiency, using continuous flow reactors' experiments. The results (Section 4.2) showed that 11.1% (v/v) oxidation efficiency was achievable for the high ratio of soil to sand mix of 9:1; while a low oxidation efficiency of 6.4% (v/v) was achieved for the low mix ratio of soil in a higher aggregate-sized sand mix of 1:9. This finding indicated that small grained-sized soil particles support more methanotrophic bacteria than higher aggregate-sized grains, due to the high specific surface area available for the bacteria to exist. The fine-grained soils can also retain methane and oxygen in the oxidation zone longer than the larger grain-sized soil can, giving more time for the bacteria to assimilate higher methane consumption efficiency. On the other hand, these fine-grained soils can clog easily and faster than can the other large-grained soils, due to the fast formation of EPS, consequently, reducing oxidation faster than the large-grained soils. This fast clogging of the filters can have an adverse effect on the oxidation process, in which an aerobic production of methane can occur, and hence, can add more methane over the already existing gas. Most important finding from these tests, however, was that having more organic matter added to the soil would help a higher oxidation efficiency. Fine-grained mix having 18.56% (wt/wt) organic matter encouraged oxidation to reach 11.1% (v/v); while low concentration of organic matter of 0.67% (wt/wt) contributed to a low oxidation efficiency of 6.4% (v/v), which is a finding that is in line with studies reached and published in the literature. From these literature studies, comparative tests were conducted on a mix of soil material with clay (Section 4.7), adding to them different mix of coconut husk of organic matter of 91.33%, concluding that these additions had caused an increase in oxidation efficiency by more than 30% (v/v) over unamended soils. Therefore, to have improved oxidation efficiencies in landfills located in arid lands, an addition of organic matter must be considered.

Also, part of the second objective of this study was to investigate the effect of oxygen penetration level into the cover layer on methane oxidation. The matter of delivering air into a medium of a landfill cover is an important aspect of methane control, and was the subject of almost all research efforts through the research community, using various amendments. The results of these efforts were found to be of mixed outcomes, because the dependency of the delivery of air through these amendments was on the atmospheric barometric and molar pressures. In this process, air can only penetrate the surface up to a maximum depth of 80 cm, with maximum presence of air at only 20–40 cm from the top medium of the cover, which was not enough to maximise oxidation. Instead of relying on these methods of soil amendments, a more efficient way of supplying air to the inside of the soil medium was to have oxygen delivered directly inside the soil medium without depending on intermediary amendments. By using a continuous flow reactor experiment (Sections 3.4.4, 4.3), it was clear, and as expected, that higher retention time of the oxygen inside the soil was supplied to interact with methane, when oxygen was made available deeper inside the soil medium allowing the methanotrophic bacteria to assimilate methane at their own pace, resulting in a maximum oxidation efficiency of 58% (v/v), compared to 49% (v/v) for surface delivery. The result also showed that with the passing of time toward the end of the experiment, and due to this higher retention of methane and air inside the medium, the oxidation reached 42.5% (v/v), compared with the surface delivery that reached only 2.32% (v/v). In the meantime, the average efficiency of methane removal from the soil during the course of the experiment reached an average of 35.66% (v/v), compared to an average efficiency of 21.53% (v/v) for the shallower oxygen delivery. This translates into more than 65% higher oxidation efficiency in favour of the deep oxygen delivery. Not only did this experiment indicate that this higher efficiency can be achieved, but also it provided a solution for air delivery inside blocked surfaces of landfill covers located in arid environments. If such deep delivery system can be devised, it can provide a venue for higher efficiencies and to can deal with the regional regulation requiring low permeability surface covers to be installed on landfill surfaces. Finally, this finding on high methane oxidation efficiency could also provide the justification for spending efforts and investments in seeking new ways and designs to

supply air deeper inside the cover layer, instead of relying on the atmospheric, molar, or the wind surface of air delivery.

The third objective of this study was to propose a system of mitigation that could reduce methane production from landfill existing in arid climates, based on accumulated data, from the literature and from experiments. Therefore, an attempt to devise a system of delivery to transport air (and oxygen) to the inside medium of the cover was done. The system consisted of a funnel and delivery pipes system, which can use the power of the wind to force air inside the oxidation soil medium, delivering the needed oxygen to the methanotrophic bacteria through a wider oxidation zone. A detail design of such system was introduced and shown in Appendix D. A numerical simulation of this system was also modelled and compared with a field trial of a bio-window filter done over Kelso Waste Depot Landfill in Sydney, Australia (Sections 4.8.1-4.8.3). The comparison between the two systems showed that for a funnel to pipe diameters' ratio of 25.89, the penetration level could reach up to more than four times that of the bio-window filter system, and could reach even deeper penetration levels for higher ratios.

The last objective of this study was to propose a scheme and recommend to the authorities in Kuwait to carry out a course of action aimed at reducing the carbon footprint resulting from landfill emissions. For the particular climate conditions existing in Kuwait, with landfills having the aforementioned poor surface vegetation, low hydraulic conductivity of the soil, and the unsuitable use of conventional mitigation systems, the information gained from the series of experiments conducted in this study can now be utilised to reduce methane emission. Methanotrophic active cover placed daily on wastes, use of common material impregnated with organic matters, and the delivery of oxygen deeper within the soils, using a funnel delivery mechanisms, can all be put together in a system to reduce LFG emission from landfills active in arid environmental conditions. Therefore, measures that must be taken to reduce some of the carbon footprint that is caused by the poorly managed landfills sites in Kuwait are suggested to be of the following:

- A small composting facility should be constructed to produce compost materials for landfills amendments, and possibly for other domestic use. Although this would be an expensive initial investment and may need some governmental subsidy at first, it is anticipated that the facility may pay off for itself, in the light of international carbon taxation, and in the event of an increase in the cost in domestic collection.
- There is the need to levy additional waste collection tax, currently at a very low rate of £25 per year per household (in a country with no income, sale, or any form of taxes), to offset some of the cost of establishing a composting facility.
- Organic matters (63%), paper discards, and discarded wooden construction materials (19%) from the approximate 500 tons of collected wastes daily in the country should be sorted and collected to be used as feed for the composting facility and as a reduction mechanism of the volumes of landfills wastes.
- Produced compost material from this proposed facility must be verified to have the optimum characteristics for methane oxidation. The material must have the qualities of being coarse, porous, organically active, and of having appropriate moisture and temperature settings.
- Daily cell covers, amended with the produced compost material having active methanotrophic bacteria, should be used and placed on top of the waste to reduce the daily methane production.
- To ensure nitrogen availability to the methanotrophic bacteria in the poor nutrient- enriched desert soil, the top covers of the landfills should be amended with the produced organic compost material.
- To ensure the availability and continuity of oxygen supply to the sand soil of low conductivity in the desert environment, an oxygen delivery system, such as the funnel system proposed in this study, should be considered, pending future field tests.

Appendix A

Landfill Covers Technologies and Methane Capture Methods

Landfill covers technologies and methane capture methods

A.1 Introduction

Disposal of wastes in ground pits and then covering them with soil have been the standard practice all over the world. With this measure of containment, it was thought that the earth would take care of the waste by itself, recycling it through its own environment, with no or little consequences. Discovering the effect of that thought on the environment, the landfill industry has since then evolved through many stages, in which it started to take account of the factors that affect water infiltration into the waste layers down into the underground water. Recently, it has become a major concern for environmental agencies. Therefore, a cover design of low permeability medium that promotes surface water runoffs, while containing erosion of the cover soil became a design requirement of any landfill site construction (US EPA regulation 40 CFR parts 258). Other design factors, such as leachate containment, gas fire prevention, and odour control were also other design factors that interested researchers, engineers, and waste managers during these early stages of landfill research. Subsequently, better landfill designs were developed for landfill covers, which included the design of liners, leachate routing and collection, prevention of excessive gas pressures and explosion, and methane gas for energy or flaring (with toxic combustion outcomes), in order to improve new landfill sites. However, it has been realized that there were other problems with landfills beside local environmental pollution. Some of these concerns were in relation to global climate changes, particularly the role of methane production from landfills in the global environmental contamination with greenhouse gases. From these problems and concerns, a cover technology field was born beginning with the wildly cited research article published by Whalen et al., in 1990, the first team to document methane oxidation emitted from landfill cover soils.

Discovering that much of methane emission was related to landfill gas production, as it has been ranked the second most polluting source after oil and gas industry sector, this had led a number of researchers to investigate and focus much of their efforts on

understanding the process of methane oxidation and the factors that affect the process. Their efforts also directed toward quantifying the rates of methane production and oxidation. The methanotrophic bacteria that was identified, as discussed in chapter I, and the cover medium that has been discovered to possess favorable general characteristics of medium—porous, good water holding capacity, stable and with a high level of organic matter— together have been identified to mitigate methane (Humer and Lechner, 1999, 2001). Thereafter, investigation of microbial methane oxidation in landfill has taken many directions, both in laboratory and field studies and is well documented in the literature , i.e., Whalen et al., 1990; Figueroa, 1993; Kightely et al., 1995; Boecks et al., 1996; Czepiel et al., 1996; Kjeldsen et al., 1997; Boerjesson et al., 1998; DeVisscher et al, 1999; Humer and Lechner, 2001; Scheutz and Kjeldsen, 2001; Huber-Humer, 2004; Gebert et al, 2005; Dever et al., 2007; Philopoulos et al., 2008; Einola et al., 2009; Bonger et al., 2010; Scheutz et al., 2011; Morris et al., 2012; Scheutz et al., 2013; Kjeldsen and Scheutz, 2014. However, the designs for methane oxidation systems are concentrated only on a set of proposed concepts (which will be discussed in the following sections), in which all researches revolved around the points of enhancing these concepts. Kjeldsen and Scheutz (2014) summarized these designed concept systems as follows: a full surface biocover, bio-window, open bed passive biofilter, closed bed biofilter, open bed, active biofilter, closed bed, biofilter, bioactive intercepting trench, and combined solutions. These systems are discussed in the following sections.

A.2 Full surface cover system

According to Huber-Humer et al. (2009), a bio-cover system is a top cover layer laid down on landfill soil to optimize the environmental conditions for methanotrophic bacteria and to enhance biotic methane consumption. A typical bio-cover is made of material that can increase oxidation and support the consumption of methane and the growth of the methanotrophic bacteria. In that process, the bacteria consume methane for energy, resulting into carbon dioxide, a substitutive and less harmful gas which is left to escape into the atmosphere. To encourage this oxidation process, a distribution layer made of a material that has the characteristic of being highly porous

is placed immediately on top of the waste body, and then a cover of uniform textured and enhancing material is placed over this distribution layer. This cover system is conceptually suggested by Huber-Humer et al. (2008) in Figure A.1.

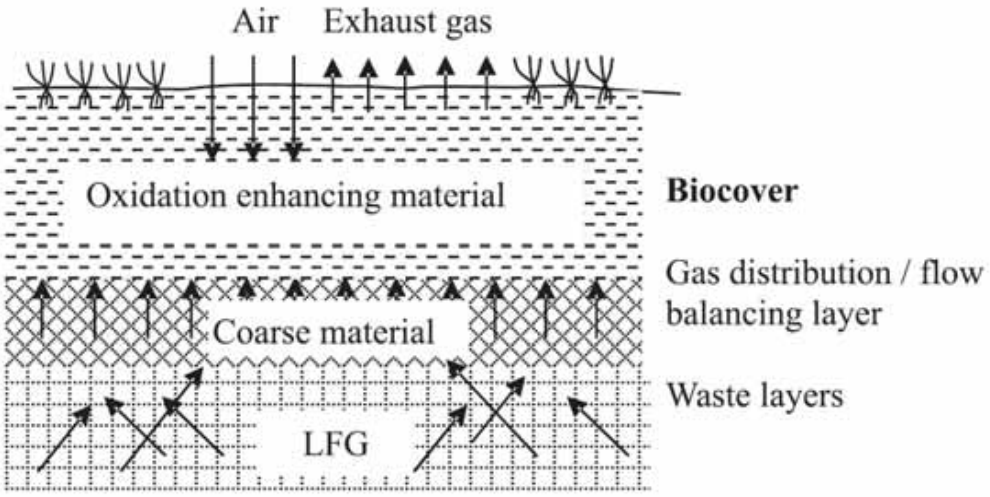


Figure A.1: Concptual design of a bio-cover system (source: Huber-Huber, 2008).

The distribution layer is often constructed from gravel, crushed glass, or ceramics of fairly coarse sizes, constructed in such a way that it allows uniform passage of the landfill gases to the bio-cover layer. The thickness of this layer is usually in the range of 10 to 30 cm (Jugnia et al., 2008) placed under the oxidation top cover layer; while the top oxidation cover layer is constructed from a material of thickness reaching up to 100 cm (Stern et al., 2007). The design of this bio-cover system is the general design concept, which is widely accepted in the literature; however, there are other design systems, but only with some variations over this design, as reported in the literature (Ettala and Vaisanen, 2001; Fones et al., 2003; Stein and Hettiaratchi, 2010; Kjeldwen et al., 2013).

Since the top oxidation layer is actually the real reactor part of the whole system which supports all the activities of oxidation, its material makeup is deemed important. The material of this oxidation cover is selected to encourage maximum oxidation and has been the subject of much research (Huber-Humer and Lechner, 1999; Hilger and Huber-

Humer, 2003; Bonger et al., 2005; Jugnia et al., 2008; Bonger et al 2010; Shangari and Agamuthu, 2012; and others), resulting in diverse results and outcomes, both, from lab works and field experimentations. Shangari and Agamuthu (2012) found that methane oxidation could account for 100% of all methane loads when using compost mixed with brewery-spent grains in a ratio of 3:7. Equally the research of Humer and Lechner (2001); Abichou et al. (2009); and Scheutz et al. (2009b), all found that 100% oxidation capacity of a bio-cover made of matured compost under certain optimum conditions, with proper layer design could be realized. In a similar vein, Berger et al. (2005) obtained oxidation rates ranging from 57 to 98%, when using a two-cover layer system comprising of a mixture of sand and compost, with one layer of 0.3-m thickness, and a second layer of loamy sand at 0.9m thickness.

A notable example of the use of compost material as a cover was the recent work of Hrad et al. (2012), in which five lysimeters (tanks systems to measure soil properties) were filled with different materials, namely, matured sewage sludge compost (SSC), matured sewage sludge compost (40% vol) mixed with sand (60% vol) as (SSC-mix), top soil and fine silt sand (TS-F), loamy soil (SS-Z), and sand soil; all were tested for 100 days. The results showed that while the mineral soils (TS-F, SS-Z, and sand) exhibited reasonable methane oxidation in the ranges of 38%, 3%, and 20%, for each, respectively, the matured sewage sludge compost showed an oxidation rate of 100% continuously for 50 days, even for higher methane loadings. Similar results were obtained by Shangari and Agamuthu (2012); Humer and Lechner (2001); Abichou et al. (2009);and Scheutz et al. (2009b).

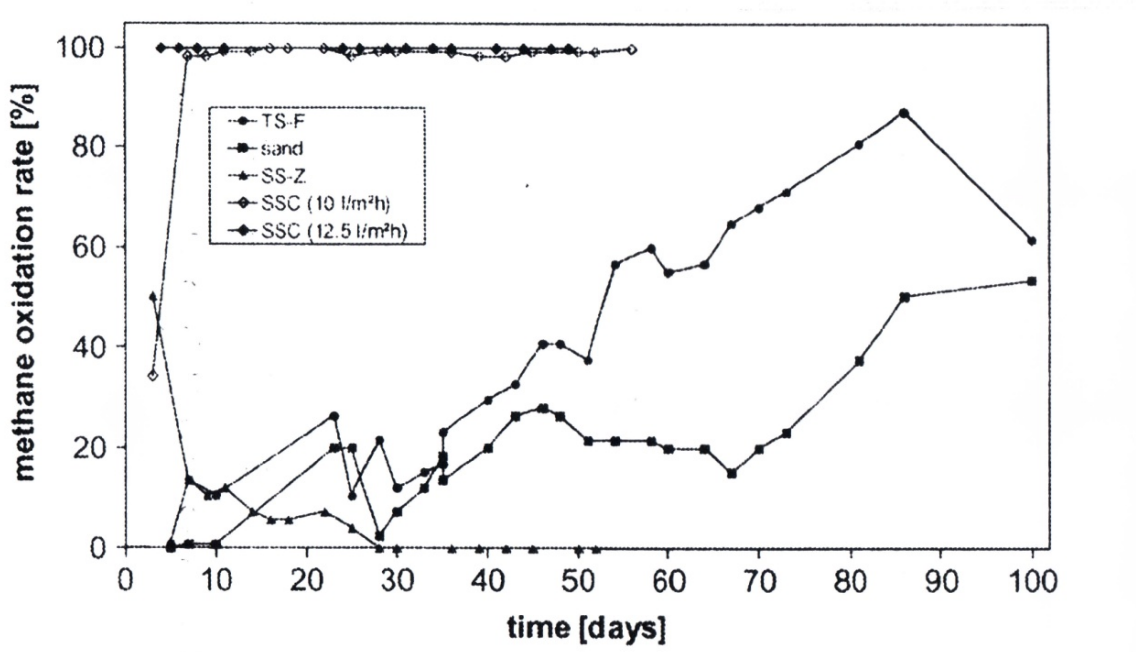


Figure A.2: Comparison of five different cover materials tested in laboratory (source: Hrad et al., 2012).

The results of Hrad et al., (2012) shown in Figure A.2 indicate that oxidation in the compost cover material proceeded steadily just a few days after the start of the experiment; while the other materials, particularly the top soil with fine silt sand (TS-F), showed gradual increase of oxidation rate, reaching up to 80% rate of methane load at 100 days. This is especially significant, because the matured compost sludge, which naturally contains organic matters and already has high level of bacteria, showed that high rate of oxidation; while the top soil material took a while to repopulate itself in the presence of methane as the source of the bacteria's energy, and then increase to reach its rate of oxidation. This would mean that a raw top soil has the potential to compete with the sludge compost cover when given enough time to adapt to its environment.

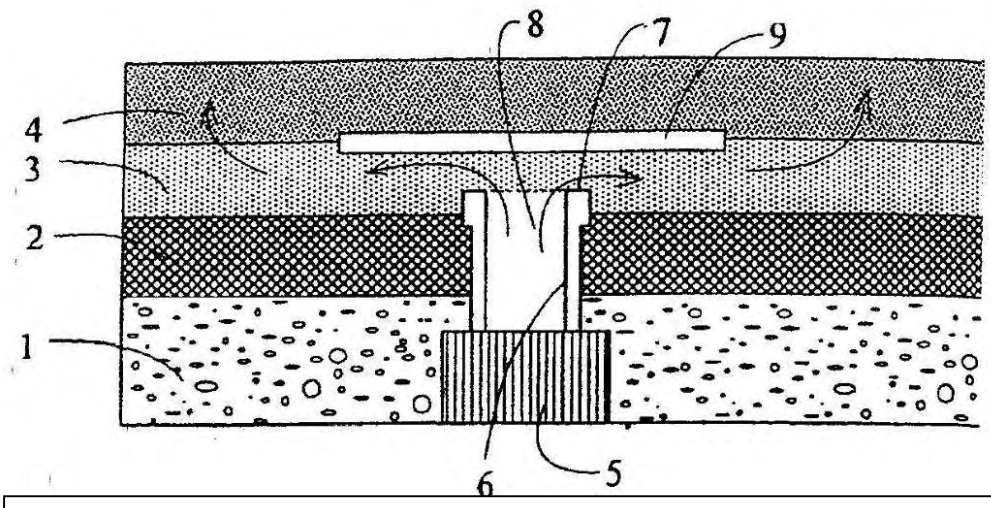
In most of the research studies in the literature, a cover layer constructed of compost material showed that if mixed with other materials would produce the most suitable cover material compared with non-compost mixes. This compost material, as indicated

by experiments, produced methane oxidation rate reaching up to 100%; although however, this material is not without its own drawbacks, as discussed in Section 2.6, Chapter II. Oxidation rates for other different cover materials will also be discussed in these subsequent sections.

Plants are an integral part of a biocover system, and can enhance methane oxidation in some instances. Plants can allow favorable conditions for methanotrophs to flourish, because penetration of their roots into the cover layer creates a support matrix for all kinds of bacteria to grow. However, plants can compete with methanotrophs for the limited oxygen in the soil, as they are themselves organic systems, hence, reducing methane uptake from the soil. Moreover, as they extend with time, their growing roots can create venues of methane to escape without being captured. In addition, some terrestrial plants can be a source of methane formation in a study by Keppler et al. (2006).

Theoretically, the importance of a biocover system in reducing methane emission is that it can oxidize up to 100% of methane, given the proper use of a cover material when it covers the whole landfill surface, so that it leaves less gas to escape. This kind of bio-cover system has low maintenance and operation costs, because of its simple design feature. However, the high permeability characteristics required of the biocover, particularly when constructed with a composted sewage, would allow significant rainwater to infiltrate through the landfill cover down to the waste material beneath it. When that rainwater is collected in the leachate layer, it would increase the contamination of the groundwater, particularly if there is no leachate collection system present. American and European regulations, at present, have mandated that landfill owners and operators should use low permeability material as a landfill-capping layer, in order to prevent hazardous gas from reaching the public and to minimize landfill gas emissions, in addition to minimizing rainwater infiltration. To resolve this characteristic problem of a high/low permeability of such bio-cover system, Ettala and Vaisanen (2001) came up with a patented system where the bio-cover can be constructed from low permeability material, as per regulations, under the highly permeable cover layer. Both of these layers are topped by a cover soil; while the methane gas can be

controlled via collection and distribution pipes, as indicated in Figure A.3. This arrangement could direct the landfill gas directly to the top bio-cover layer, while preventing rainwater infiltration into the waste layers.



1: Refuse 2: Impermeable layer 3: Drainage layer 4: Top soil cover layer
5: Gas collection pipe 6-8: Distribution well placed through impermeable layer
9: Impermeable layer to prevent gas immediate escape.

Figure A.3: Patented bio-cover design system (source:Ettala and Vaisanen 2001).

There are more unfavorable characteristics inherent on the use of a complete biocover material over landfills. Even though the covering of the entire landfill site with a biocover is desirable to encourage optimum oxidation, and possibly to extend the coverage to behind the perimeter of the landfill itself, the amount of material required as a distribution layer. Gravel or similar materials if used to cover the entire landfill surface can be quite expensive, particularly if such materials are not available in the region. This unavailability in the region would require transportation from other regions and distribution efforts, to produce greenhouse gases. Such regions that have no gravel base or gravel sediments include Kuwait and many other desert regions, where the soil throughout the country is only hardpan calcareous soil. In addition, the top bio-cover layer, when made of composted material, has its own problems. The composting process by itself produces high amount of greenhouse gases, and when transported and distributed over landfill surface, would again, add more greenhouse

gas to the environment. Furthermore, compost materials must be matured, in order for methane to be optimally oxidized (Schuetz et al., 2011). However, reaching that maturity stage of composting requires an aeration of the compost for extended periods of times in order to eliminate anaerobic production of methane from the raw compost, a process of releasing methane from composts to the atmosphere; then, the material itself is used again to capture the methane gas from the cover layer on a landfill, as one of its drawbacks. Furthermore, the availability of compost venders in some countries and the availability of compost material for field scale application is limited all year round, which makes it less preferred by landfill engineers and operators. In addition, compost cover material, with time, will experience settlements, reaching up to 20% of its original volume (Schuetz et al., 2009), therefore reducing its permeability potential, and hence, its capacity to oxidize methane. Moreover, compost material could develop cracks and voids during settlement that create preferential flow path for landfill gases, and will short circuit the methane flow directly to the atmosphere without oxidation (Chanton et al., 2011). The final issue with biocover systems is the inability to control the rate of oxidation process and provide a homogeneous and equally effective cover layer (Huber-Humer, et al., 2008). All these drawbacks of a biocover system have prompted researchers to investigate other alternatives to covering the whole landfill site, and investigated other systems such as the biofilters, biowindows, and the biotarps.

A.3 Biofilter system

Biofilters are another systems used for methane control that could be installed passively or actively. In any of these designs, a suitable oxidation layer is placed over a distribution layer, and unlike the bio-cover system, all layers are contained in an enclosure for the purpose of avoiding the disadvantage of the bio-cover system, which was the lack of control over the oxidation process. The system is placed inside the landfill structure, or above the capping layer. In this contained system, the main variables, such as methane loadings, moisture contents, air inlets, and the material makeup of the filter can be readily controlled.

A.3.1 *Passive biofilter systems: Open bed*

Passive biofilters are constructed and used in many ways. Three of the earliest biofilter design types used for methane mitigation were the ones introduced by Straka et al. (1999). They used them in landfill gas elimination process in four old landfill sites in Czech Republic. Three design types were used, namely, the pile type, placed above landfill ground, the middle sunk, partially positioned inside the top soil of the landfill, and the countersunk, a biofilter totally immersed inside the top soil, as indicated in Figure A.4.

In these proposed systems, a container of geotextile barrier is placed inside the top soil of the landfill above, middle, or completely sunk inside the soil, filled with a compost oxidation material as a bed, erected open to the atmosphere and placed over coke distribution material. The designed systems were fed passively with landfill gas from the bottom of the containers in an upflow orientation via perforated pipes. The systems are designed with a leachate collection trench constructed on the bottom of the biofilter, and extended away from the container.

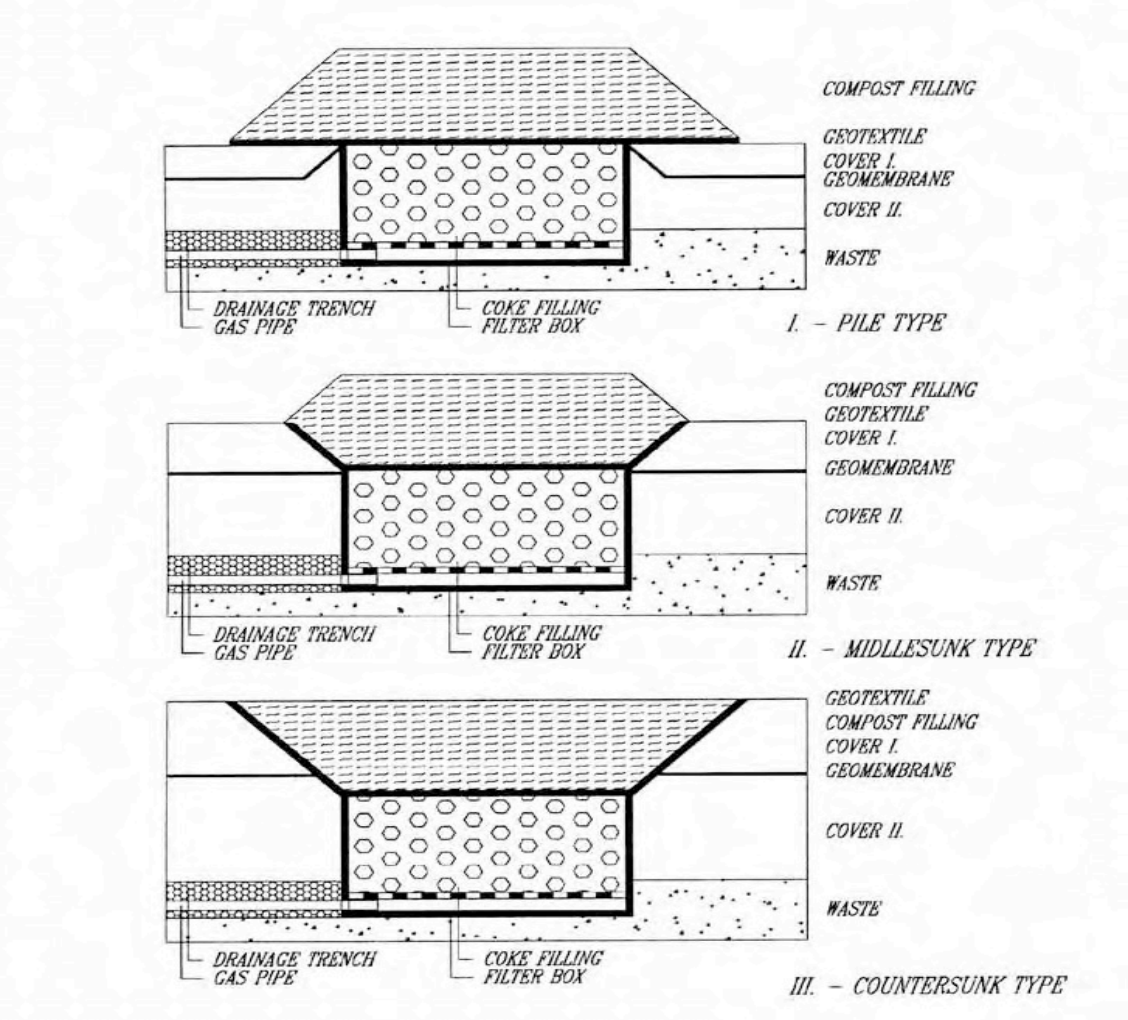


Figure A.4: Three open bed types of biofilters proposed by Straka et al. (1999).

In their observations of their systems, Straka et al. (1999) achieved an oxidation rate reaching up to 90% v/v with a methane loading of 26.4–60 $m^3\ m^{-3}d^{-1}$, when compost layers of 0.8- to 2.0- m thickness were placed inside the biofilters. They also observed that monitored methane emission from the filters was generally less at 0.1% of raw methane makeup of 55% from the landfill. Finally, the Straka et al. (1999) group had concluded from their investigations that biofiltration systems are much cheaper than a collection of incineration operations.

There have been a few investigations on the operations of biofilters in field experiments, besides the one done by Straka et al. (1999). Gebert and Grongroft (2006) investibated an open bed biofilter containing porous clay pellets as a

distribution layer, and placed under an oxidation layer of topsoil material, the system being fed passively in an upflow orientation (landfill gas fed from below) with landfill gas containing up to 55% v/v methane concentration. The methane load was 5928-g CH_4 m^{-2} d^{-1}, which resulted in high oxidation rate of 1920- g CH_4 m^{-2} d^{-1}. They also reported a notable conclusion, stating that, when methane loading was increased, the rate of oxidation dropped, due to limiting oxygen diffusion into the bed material for the prevailing high flow of methane.

A.3.2 *Passive biofilter systems: Closed bed*

Closed bed passive biofilters are similar in design to the ones proposed by Straka et al. (1999), as in Figure A.4; however, they differed in that, the filters are closed in a tightly confined containers and shielded from the atmosphere. In this system, landfill gas and oxygen are fed passively into the filter, either by mixing or by separate feeds to the biofilter, as indicated in Figure A.5 (Schuetz et al., 2009).

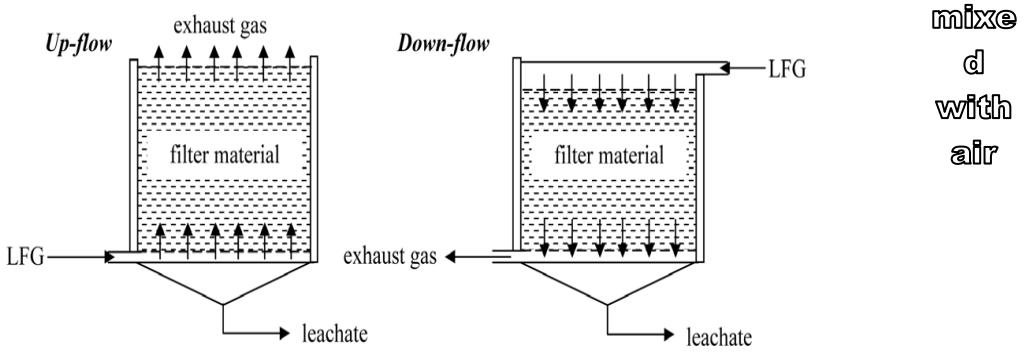


Figure A.5: (a) Open bed upflow and (b) closed bed downflow biofilter systems (source: Schuetz et al., 2009).

This system of biofiltration has been tried in field trials by Powelson et al. (2006) in which two closed bed biofilters were used; one was contained a mixture of chipped

yard waste compost and polystyrene pellets; while the other had a combination of different grain size sands. The design of the second biofilter had graded sand grain sizes from fine at the top to coarse size at the bottom, to provide optimal moisture and air at the some point within the bed material. When the two systems were fed with synthetic LFG mixture of methane of 55% v/v with flux reaching up to 750-g CH_4 m^{-2} d^{-1}, the two filters were found to perform similarly with 63% and 69% of the methane being oxidized by the sand and compost filters, respectively. These results indicates that a sand filter, when supplied with sufficient oxygen to penetrate the sand, and promoting oxygen diffusion, can performed as well as a compost material filter. This notable results, indicating that oxygen supply and penetration deep down into the bed layer to supply the methanotrophic bacteria with its needs of oxygen are more important than the type of material used as a packing material.

Closed bed biofilters are useful, not only for continuous methane reduction in landfill sites, but also because they can be used in a variety of ways. One way of usage is to employ the closed bed filters as a temporary means to reduce the amount of gases from escaping the ongoing landfill operations. In this operation, the biofilter system is placed on one section of a landfill until it is sealed; then, moved again for every time a landfill section is sealed further onto the following sections that are being filled. In this setup, the gas is mixed with air and fed directly into the filter to oxidize the gases that are temporarily escaping from the site; while it is in active operation. Another way is to employ such filters in reducing contaminated gas and the treatment and control of odours emanating from industrial operations, such as sewage treatment plants, composting operations, poultry industrial waste, etc., employing the same principle as previously discussed (Figueroa, 1996).

A.3.3 ***Active biofilter system: Open bed***

Active open bed biofilters are designed in the same manner as the closed system, except that air and pollution gases are pumped into the system in an upflow or downflow directions in an active operation. For this type of a system, the resulting gases, as an output, are vented directly into the atmosphere in a similar concept, as

shown in Figure A.6 (Leson and Winer, 1991). These systems of pollution control have been successfully used in many industries that have low pollution concentrations, such as in sewage treatment plants, food industries, chemical plants, incineration plants, agricultural processing industries, animal breeding industries, composting facilities, etc. They are most widely used, however, to deodorize and control odour of waste treatment plants and animal breeding facilities (Ando, 1980; Bohn, 1975; Prokop and Bohn, 1985; Leson and Winer, 1991; Schigeimllch, et al., 2005).

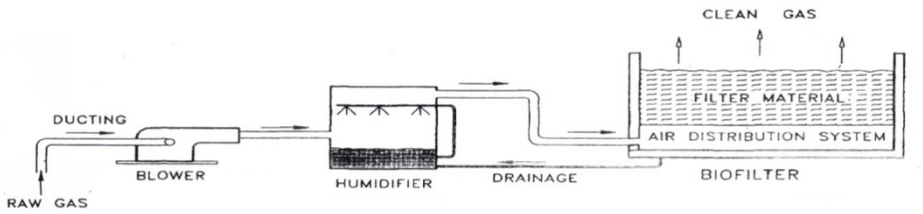


Figure A.6: Open bed biofilter, actively fed with gas and air (source: Gero and Winer, 1991).

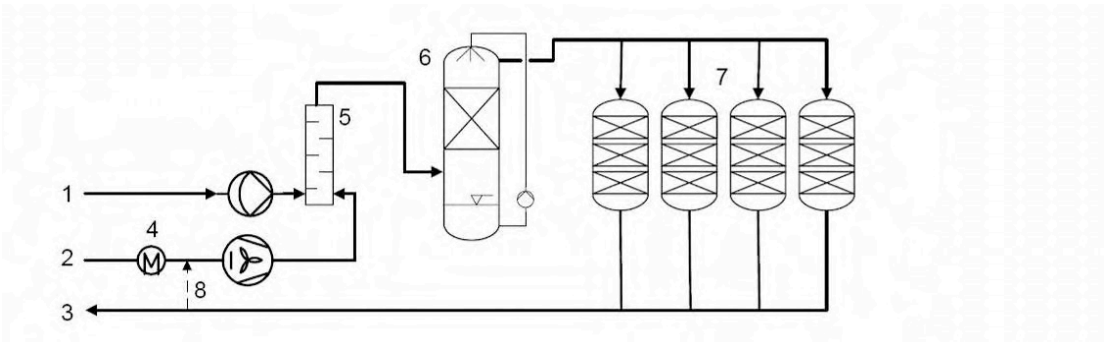
In a field study by Gero and Winer (1991), using open bed active biofilters to reduce volatile organic compounds (VOC) from industrial raw off-gas, it was shown that biofilters are most suitable for low VOC and can reduce ethyle ocetate, butyle ocetate, and toluene up to 30, 35, and 25-g (VOC) m^{-3} h^{-1}, respectively, for raw gas loading of 275 m^3 m^{-2} h^{-1}. They also indicated that if several organic compounds were present, their degradation capacities may behave synergistically, resulting in higher total degradation rates than if each compound were only present alone. In another field study dedicated to food production industry toward the poultry production, specifically, Tymczyna et al. (2004) used an open bed biofilter to assess the removal efficiency of biofiltration of chemical and biological air contaminants from vented gas of a laying hen house. The biofilter bed was composed of 35% fibrous peat, 35% sallow peat, 10% barley straws, and topped with composts from sewage plant and horse manure. They reported that ammonia biofiltration efficiency ranged from 36 to 89%,

and nitrates, nitrites, and phosphates uptake rates were much higher reaching up to 66–100%.

Field studies regarding methane removal, using open bed active biofiltration systems, are scarce in the literature. The reason for this lack of studies could be attributed to the relative cost involved in using this type of pollution removal method, compared to the advantageous properties of using open bed passive system, or the bio-window system. It also could be attributed to the lack of control over the exhaust gases escaping to the atmosphere as a result of this type of biofiltration.

A.3.4 *Active biofilter systems: Closed bed*

Closed bed biofiltration system is different from the open bed system in that, it is completely enclosed and shielded from the outside elements so that reaction conditions controlled. The system consists of a biologically active bed, or several levels of active beds, stacked together for maximum efficiency, a possible distribution layer, raw gas feed, and air input; all are controlled by feed pumps and humidification shown in Figure A.7 (Streese and Stegmann, 2003).



1. Landfill gas. 2. Air. 3. Exhaust. 4. Air heater. 5. Mixing chamber. 6. Air humidifier. 7. Biofilters stacks of 12 in 4 columns. 8. Recirculation line.

Figure A.7: Pilot scale plant with several filters constructed by Streese and Stegmann (2003).

Functional control of active closed bed extraction biofilters and ease of operation have been investigated by a number of researchers under different conditions and materials types (Streese and Stegmann, 2005; Nikiema et al., 2005, 2007). In their investigations, Streese and Stegmann (2003) constructed a pilot scale plant with several filters to test a variety of bed media, which included matured yard waste compost, peat, spruce wood fibers, and yard waste, and experimented with them as mix or as an individual. From their investigations, they have achieved an oxidation rate of up to 1488-g CH_4 m^{-3} d^{-1}. They concluded that active biofiltration extraction systems have the potential to eliminate landfill gases even though they have relatively low rate of extraction compared to odour elimination biofilters, and that it would require a large-sized extraction system to be more effective. However, even with that size the costs of construction and operation of such system were expected to be low (Streese and Stegmann, 2005). They concluded further that active biofilters are cheaper to operate at 30°C than at 22°C, with respect to initial investments and operational costs.

Naturally, in this kind of active extraction, the oxidation rate is much higher than that of a passive system of comparable size. This is due in part, to the ability of operators to control air into the oxidizing reactor bed to be mixed with the landfill gas for desired oxidation efficiencies, however, at a cost and comes with added operational difficulties. Such difficulties arise from the formation and clogging of the biofilters media by the EPSs, which reduced the performance substantially (Dammann et al., 1999; Streese et al., 2001, 2003, 2005). It is worthy to note that such a system had higher rates of oxidation than other passive systems, because when oxygen was made available and penetrated well into the reactor bed, reaching more methanotroph aggregations, the rates of oxidation had risen substantially in relation to any passive systems. If such a system of having low costs of operations and high performance could be had and could be maintained operationally, then that system would be an optimum solution and would be most effective in methane elimination. Similarly, Slezak et al. (2015) have also concluded in their investigation of a simulated landfill containing municipal solid wastes, using lysimeters containers, that the aerated systems have the potential of oxidation 5 times more than an aerobically tested system.

A.4 Bio-window system

Unlike the bio-cover or the biofilter systems, where the whole surface of the entire landfill site is covered with oxidation-enhancing soils, for the first, or contained in a controlled environment in the second, a bio-window system is a system designed to be installed in discrete parts of the surface of a landfill, preferably right on the hot methane emission spots. The system is installed without a barrier container. In these hot spots of closed or old landfills, where methane emission is detected in the cover surface, bio-windows could be most suitable to mitigate methane emission from the whole landfill by placing them in specified places. These bio-windows are constructed in the landfill cover soil without containments and without support structures, therefore much cheaper to operate. The top part of the bio-windows is often made of biologically active material such as compost, placed directly over the waste layer of the landfill. A conceptual design of bio-windows was first introduced by Huber-Humer et al. (2008), as shown in Figure A.8.

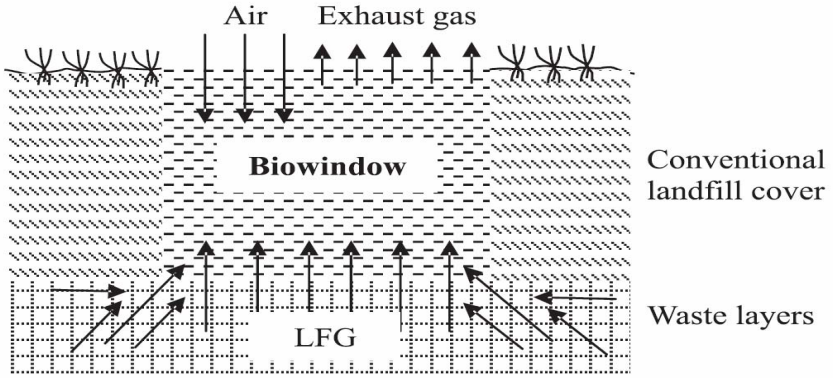


Figure A.8: A conceptual design of a bio-window (source: Huber-Humer et al., 2008).

In this passively designed system, the bio-window bed receives methane-form waste from the bottom, in a way of advective flux driven by pressure gradients, and oxygen from air, induced by way of diffusion from the top of the cover driven by molecular gradient, or advection by wind or atmospheric gradients. A field trial experiment to test the performance of this kind of a system was conducted by Scheutz et al. (2011) on Fakse landfill in Denmark, in which a bio-bed of garden waste compost was imbedded

into a clay cover. After monitoring for approximately 400 days, they obtained a 41% oxidation efficiency rate and a 28% methane emission reduction from the landfill, which showed lower oxidation efficiency than a full bio-cover system or an active biofilter system. These lower rates are naturally expected, since the bio-window system is in actuality a partial bio-cover system, in which only a portion of the top cover of the landfill was used. Additionally, oxygen delivery mechanism depends only on the natural elements of the atmosphere; however, the capital and operating costs and the easiness of operations are very favorable for this system, particularly for the mitigation of landfill gases in landfills that do not have an active extraction system.

A.5 Bio-tarp system

Substantial methane emission can escape from landfills during the active filling period; therefore, a measure to combat this escape during this phase of filling is required. This measure comes in a way of a cover laid over daily on the cell that is being filled at a landfill site as a temporary solution to combat odour, flying windblown debris from the waste, scavenging animals, flies, and to prevent flash fires, in addition to mitigating landfill gases. The bio-tarp can be made of materials such as sewage sludge, yard waste, or a commercially available geotextile material impregnated with methanotrophic bacteria. If these materials were not available, a 15-cm layer of soil can be laid daily over the dumped wastes, which is the method used mostly in practice (Huber-Humer et al., 2008). However, covers such as the geotextile cover material, when used and removed daily as a temporary cover, can save space and time for use in further filing of wastes on the site. The tarps are placed at the end of the day and removed the next day for further filling, then placed over again at the end of that day, and so on, for the time it takes to complete filling the site space, which can take a long time. During this long period, a geotextile cover material impregnated with methanotrophs can effectively mitigate the emission of methane from the landfill. A conceptual operational bio-tarp system is shown in Figure A.9. In a study to quantify the effectiveness of this kind of a cover, Adams et al. (2011) found from their investigation that this system can remove 16% of methane, and when combining compost material with the geotextile cover, the oxidation rate could double to 32%.

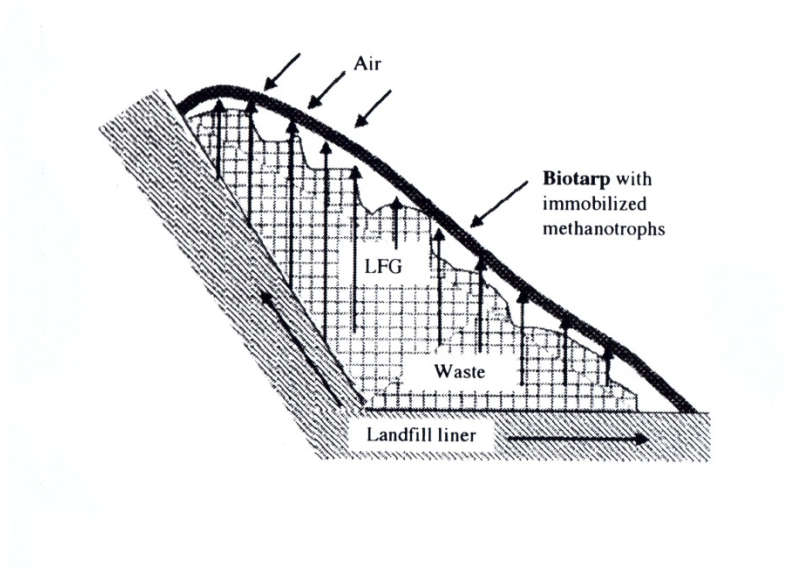


Figure A.9: Placement of a bio-tarp over partially filled landfill site (source: Huber-Humer et al., 2008).

One of the advantages of the temporary bio-tarp is that it can be recharged with methane, consuming bacteria every so often, that is, when physical or environmental conditions do not degrade this temporary cover, bacteria are reduced or removed. Much like the other bio-covers, the bio-tarps systems are affected by the same factors that disturb the other systems, such as moisture content, temperature, porosity, etc. However, a continually removable tarp must possess properties of good moisture holding capacity, porosity, durability, and should be of light weight to maintain continuous transport. Unfortunately, all these properties are hard to obtain in one single material. The other concern relative to bio-tarp is that, it has little capacity to remove methane from landfills, which does not exceed more than 16% when using geotextile material for its makeup, as should be the case for the most appropriate tarp material. Nevertheless, the bio-tarp is useful for temporary measures, as well as for use to prevent odours, scavenging animals, flies, and fires.

A.6 Bioactive intercepting trench and combined system

Much like the biofilter systems as already discussed, the bioactive intercepting trench systems have the same conceptual design as the biofilters in every aspect of the design, including a flux distribution layer and a bio-support medium cover, except that

it differs in the extent of the coverage area and its location in a landfill site, which are much wider. While biofilters are constructed at specific areas in marked locations on the landfill surface, the bio-trench is constructed on the perimeter of the landfill of appropriate width and depths for the purpose of catching lateral fugitive landfill gas, and consequently, to protect surrounding underground water and to prevent gases from reaching close by basements of buildings. In a field investigation study on the effects of lateral gas transport from a landfill in Denmark (Skellingsted landfill) to adjacent soils, Christophersen et al. (2001) have found that landfill gases are present in the adjacent soil; however, they are seasonally dependent. They observed that high levels of carbon dioxide emission combined with low levels of methane in the summer are influenced mostly by cover moisture levels and by the distance from the landfill site. They concluded from their investigation that the high rate of carbon dioxide at the perimeter of the landfill, estimated to reach up to 89%, is due to the high oxidation rate of methane at the landfill. The low rate of lateral methane transport could be due to the low rate of methane generation at that the old under investigation.

Very little research has been conducted to quantify the effectiveness of bioactive intercepting trenches to mitigate landfill gas. The reason for this could be due to the difficulty of quantifying the amount of lateral landfill gases seeping from the landfills to the surrounding areas and to the costs of constructing such a system for testing.

Another biofiltration design concept, proposed and tried, is the aerated passive biofiltration system. The concept is operational rather than an actual difference in design concept. The system is practically a passive biofilter concept with the same concept design as those discussed before (open or closed), constructed from a distribution layer and topped with a bioactive oxidation layer in a container, with one operational difference, which is the introduction of a continually circulating stream of air into the inside of the biofilter bed. This concept was proposed to counter the problem of high methane purging of oxygen from the biofilter's bed, when methane flow rates exceed the quantity of oxygen supplied by diffusion of the atmospheric air into the filter, and limited oxygen supply to the methanotrophic bacteria. Therefore, a counter aeration of the biofilter's bed could provide the necessary supply of oxygen to

the oxidation layer. To test this concept, Haubrichs and Widmann (2006) investigated the effect of aeration on a passive biofiltration system in a laboratory experiment, in which oxygen was provided continuously through the layers of the biofilter. The experiment revealed that in an aerated biofilter, the oxidation rate had increased to a significantly higher rate of 28.8-g CH_4 m^{-3} d^{-1}, compared with the un-aerated biofilter system of the same settings, which had only 5.1-g CH_4 m^{-3} d^{-1}, when the experiments were run for 148 days. This result indicated that when oxygen was delivered to the full bed of the biofilter, supplying far more volume of oxygen to the methanotrophic groups, the oxidation rate had naturally increased substantially, however, the fast formation of EPS and the drying conditions of the biofilter's bed were identified as problems yet to be resolved.

Other bio-mitigation systems suggested were a combination of any of the aforementioned systems in a one set of solution for methane removal. A good example is to construct a system, consisting of a full surface bio-cover bed to capture fugitive upward moving gases, combined with a bioactive intercepting trench system to capture lateral fugitive gases, or a full bio-cover system with a biofiltration system connected to a gas extraction system (Kjeldsen and Scheutz, 2014). This would cover most roots of methane escaping from the landfill. However, such a combination system would be prohibitively expensive.

To increase the methanotrophic activities in landfills, conceptual systems such as the ones discussed are required, particularly when these concepts are used for old or small landfills, or when the installation of methane gas systems is unavailable, or when these concepts are used as complementary to gas collection systems. However, the characteristics of the oxidation medium, as a bed for methanotrophs to grow and function efficiently, are just as important. Material characteristics, other than top soil covers, can be manipulated and used as amendments to the cover soils in order to create favorable environment for microbial activity.

Appendix B
(Additional Experimental Data)

Effects of inhibiting substances on the methane oxidation rates and additional data

The last test in this series of investigations was to investigate inhibiting substances that may have an effect on the rate of oxidation and on the bacteria existence in the soil samples. Therefore, a 0.25 ml of Bromoethane-Sulfonic-Acid (BESA), a strong inhibiting solution was added to samples Land-MCP (in duplicates as Samples 5,6), Land-LCP, (Samples 7) and Pure culture (Sample 10) having volumes of 1ml each, then mixed with 10 ml nutrient solution, as indicated in Table B-1. The samples were all placed in sterilized bottle reactors, 70:30 air to methane volumes were maintained, and placed on bench for measurements. The samples were also subjected to the same external condition as in the other batch samples tested previously in the Sections 3.3.1-.3.3.4. Syringe gas samplings and equipment used in this test was handled in the same ways as in the previous tests.

Sample BES 5	1 ml of	Landfill top soil samples taken from different locations (Coxhoe Lnadfill, Newcastle , UK	in 10 ml of media solution	plus 0.25 ml of BESA
Sample BES 6				
Sample BES 7				
Sample BES 10		Pure Methanotrophic culture		

Table B-1: Some Batch samples of Table 3.5 with added Bromoethane-Sulfonic-Acid (BESA) as an inhibiting solution.

In this final test, where the inhibiting substance of 0.25 ml of BESA was added to the samples as described in Table B-1, the behavioural characteristic of the bacteria under these circumstances was sought. The effect of inhibition is clear, and is shown in plot C-1. Adding BESA to the samples, taken from the landfill and a laboratory grown pure methanotrophic culture, was intended to limit the effect of methane producing bacteria by inhibiting the presence of methanogens, present in the soil. If the

elimination of methane producing bacteria is successful, then only methanotrophic bacteria would be able to oxidize methane more rapidly. The result of this addition on the samples is shown in Figure B-1, where it seemed that the inhibiting substance had also an effect on the methanotrphic bacteria as well. The methane percentage did not change for the landfill samples for the first week; however, it started to increase rapidly for the second week for the landfill samples and for the pure culture sample. This clearly indicates that some bacterial action was there, producing some methane, raising the level from the original 22% v/v to 31% v/v in two weeks. This is in contrast to the samples tested in Figures 4.6-4.8, where oxidation was almost immediate. However, after two weeks, methane elimination commenced rapidly, indicating that the bacteria had biologically overcome the inhibition effect and methanotrophic bacteria started to assimilate methane. Two observation can be noticed from this experiment, first, that the inhibiting substance effect is not clear on the bacterial community in the samples, and second, that the pure culture sample behaved differently from the naturally existing bacteria. This behaviror is expected, since the genetic makeup of the laboratory is not exactly the same as the naturally existing bacteria in nature. No further investigation was sought for this type of investigation, since that needs special require microanalysis, and special equipments.

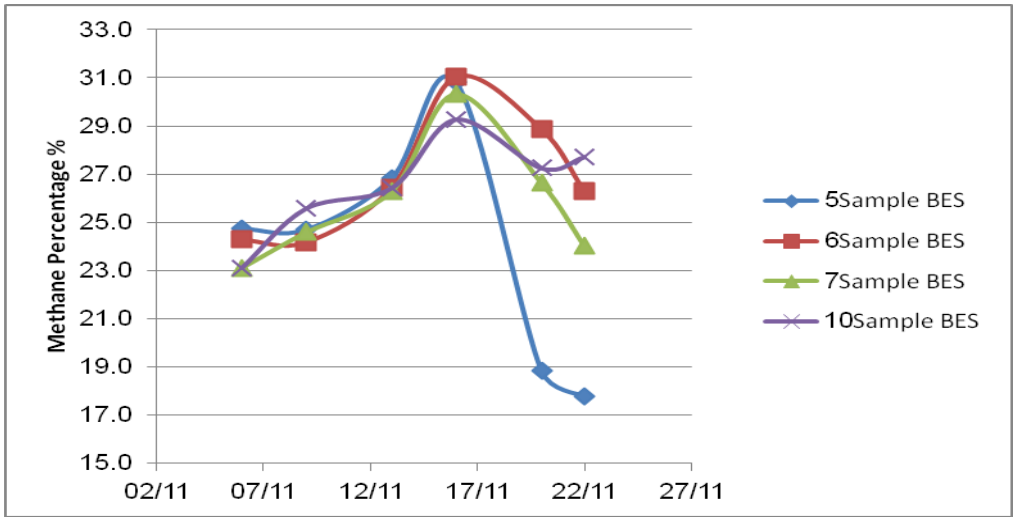


Figure B-1: The effect of an inhibitor (BESA) on the soil samples of Table B-1, showing stronger inhibition at the beginning and low inhibition with passing of time, indicating strong propensity of the methanotrophic bacteria to assimilate methane.

Additional samples under different conditions

Sample 1a	Shaker	Raw Landfill Soil Sample
Sample 1b		
Sample 2a		Soil with Sand
Sample 2b		
Sample 3a		Soil with D. Water
Sample 3b		
Sample 4a		Soil with Nutrients
Sample 4b		
Sample 6a	No Shaking	Soil Sample Raw Landfill
Sample 6b		
Sample 7a		Soil with Sand
Sample 7b		
Sample 8a		Soil with D. Water
Sample 8b		
Sample 9a		Soil with Nutrients
Sample 9b		

Table B-2: Soil samples subjected to different additives, prepared for placing on still and shaking platforms

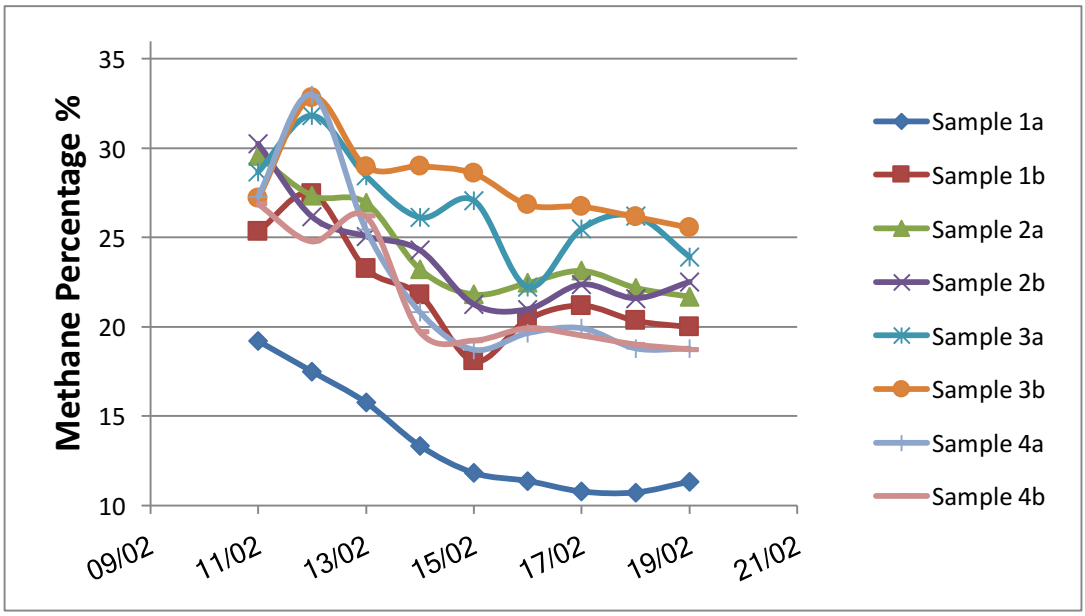


Figure B-2: Soil samples of Table B-1 under shaking conditions.

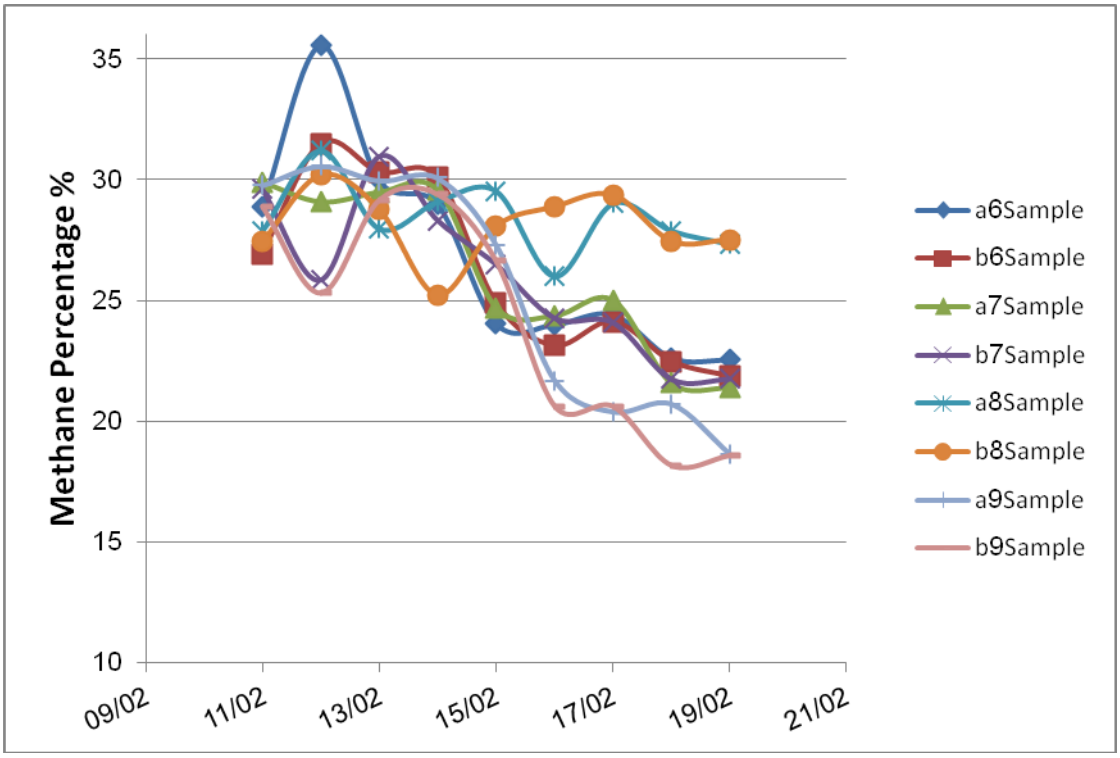


Figure B-2: Soil samples of Table B-1 under still (non-shaking) conditions.

Sample 1a	Shaker	Soil Sample with neutrients♂Raw Landfill
Sample 1b		
Sample 2a		Soil with Sand with nutrients
Sample 2b		
Sample 3a		Soil with D. Water with nutrients
Sample 3b		
Sample 4a		Soil with Nutrients
Sample 4b		
Sample 6a	No Shaking	Raw Landfill Soil Sample with nutrients
Sample 6b		
Sample 7a		Soil with Sand with nutrients
Sample 7b		
Sample 8a		Soil with D. Water with nutrients
Sample 8b		
Sample 9a		Soil with Nutrients
Sample 9b		

Table B-2: Samples of table 5.2 with added nutrients.

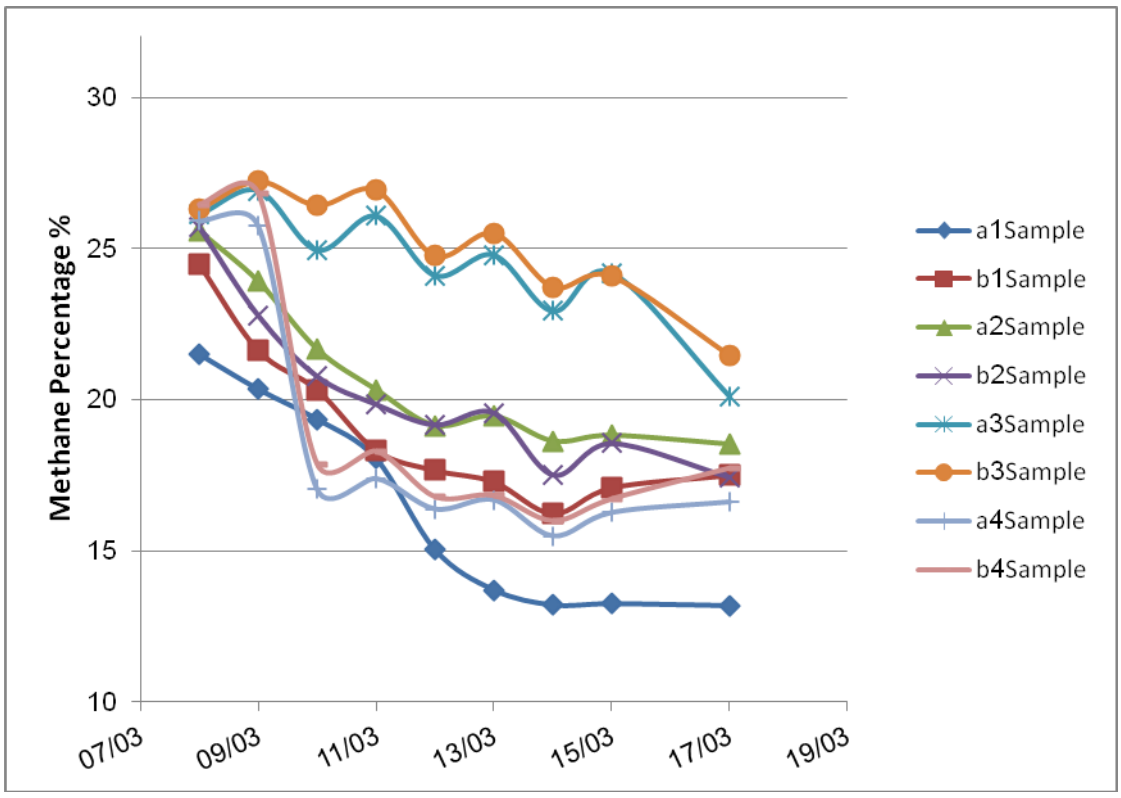


Figure B-3: Soil samples of Table B-2 under the shaking conditions.

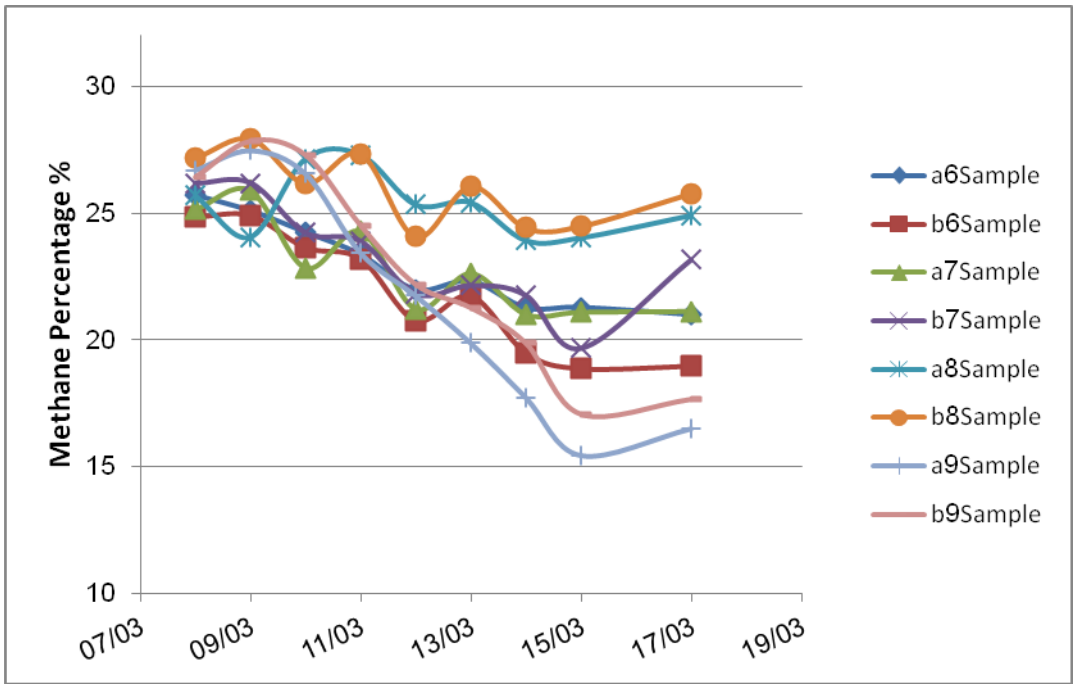


Figure B-4: Soil samples of Table B-2 under still non-shaking conditions.

	Salt solution (g/liter)	Phosphate solution (g/liter)	Trace metal solution (mg/liter)	Iron solution (g/liter)	Sulfuric Acid
Solutions	$NaNO_3$ 85 K_2So_4 17 $MgSO_4$-$7H_2O$ 3.7 $CaCl_2$-$2H_2O$ 0.7	KH_2PO_4 53.0 NO_2HPO_4 86.0	$ZnSO_4$-$7H_2O$ 288.0 $MnSO_4$-$7H_2O$ 233.0 H_3BO_3 62.0	$FeSO_4$-$7H_2O$	H_2SO_4
Amount	100	100	500	1000	5ml/100ml Iron solution

Table B-3: Commercially available media solution for cultures used in batch experiment

Source:

Brian G. Fox, Wayne Afroland, David R. Jollie, John D. Lipscomb, [31] Methane monooxygenase from Methylosinus trichosporium OB3b, Methods in Enzymology, Academic Press, 1990, Volume 188, Pages 191-202, ISSN 0076-6879, ISBN 9780121820893, http://dx.doi.org/10.1016/0076-6879(90)88033-7.
(http://www.sciencedirect.com/science/article/pii/0076687990880337)

Appendix C
Experimental Risk Assessment

Risk Assessment

Project title: Methanotrophic Reactive Barrier System for Suppressing Fugitive Gas Release (Civil Engineering and Geoscience school, PhD degree)

Student: Eng. Abdulaziz Al-Shareedah

Supervisors: Dr. Paul Sallis and Prof. David Graham

University: Newcastle University, UK

Sponsor: Kuwait Institute for Scientific Research (Scholarship)

Introduction

The research project will study the relationship between methanogenic bacteria in order to utilize it in an engineered layer that uses methanotrophic microorganisms as a barrier to reduce or eliminate greenhouse gases such as methane more efficiently and the layer can be implemented on top during the landfill operation or even after.

The study will be prepared in the laboratory on a small scale using many reactors to simulate real situations and by analysing the methane oxidation by methanotrophs and experimenting with different variables to achieve and understand the optimal oxidation rate for different circumstances. Many chemical, physical, and biological factors affect methane oxidation, some are well investigated and some need further examination, therefor more research will be processed in order to utilize methanotrophs in the optimized barrier solution. The focus will be on enhancing microorganisms media and the affecting variables such as soil types, chemical and physical characteristic, moister content, also other engineered synthetic kinds of media to develop a medium with ideal chemical and physical characteristic for methanotrophs to thrive.

The outcome of the study will be beneficial to reducing greenhouse gases by having a solution for non-sanitary landfill, even though modern sanitary landfills have a complete gas collection system that collects gases used as a source of energy. However, in less developed countries, gas collection systems and maintenance are expensive, thus, neither practical nor feasible. Similarly, it may not also be practical for those old closed landfills that are not sanitary, as well as for modern sanitary landfills that have faults from natural disaster or have poor implementation.

Experiment hazards

In order to accomplish our research project many laboratory experiments will take place, the main experiment will be focused on working with methane gas and anaerobic-methanotrophic bacteria which both are naturally available in the environment and both do not have direct health risks. The only hazard is with the methane gas which is flammable, so safety procedure to contain the gas, to dispose the gas and prevent fire must be implemented.

Safety measures and controls

The main safety focus is to prevent fire from the methane gas accumulation:

1- The experiment setup (see option-1 in Fig.1 and option-2 in Fig. 2) starts with a methane gas cylinder to simulate methane from regular sources such as landfills and an air cylinder; the cylinders will be handled by laboratory professionals and will be fixed in the laboratory as all gas cylinders to prevent it from falling.
2- All cylinders have on/off valve switches to close or open the gas flow, and also will be fitted with gas regulators to regulate the gas pressure.
3- For added safety measures rather than using pure methane thru the reactors we will mix air with the gas cylinder flow which will reduce methane concentration to 20% maximum concentration.
4- Most experiments will have more air flow added to mix with the methane gas flow to reduce the total methane concentration going to the reactor (10%, 5%, 1% methane concentration), for the first option the mixing will be at the beginning rather than mixing inside the reactor as the second option.
5- The system will be set to have a low flow (about 5-10 ml/min), so even if there was a line brake or leak, the low flow with the low concentration of methane will not fill or affect the laboratory and would be minimal. (see scenario calculation)
6- A flow meter will be placed before the reactor to measure the gas flow and to make sure the flow is constant and within the required measures, also if any leaks develop flow drops will be noticed.

7- The reactors size will be small and filled with media (such as soil) so methane amounts will be small and no methane accumulation can occur, also there is no ignition source inside the reactor.
8- Gas outlet will be vented to the atmosphere thru laboratory gas vents (fan extractors).
9- System tests will be done in the beginning with only air to check the system for any leaks or weak areas that needs to be addressed.
10- Continues leak checks will be done when the system is in use, first method is by adding known amount of pressured gas in the system pipes and closing it and leaving it for couple of day to check if there is a pressure drop, the second method is by using soap water and applying it on the pipes to see any gas bubbles forming.
11- Signs will be placed on the laboratory doors to indicate flammable gas cylinders are present, and risk assessment documents with safety procedures will be placed besides the reactors.
12- The gas cylinder will be closed from the source when the experiment is stopped.

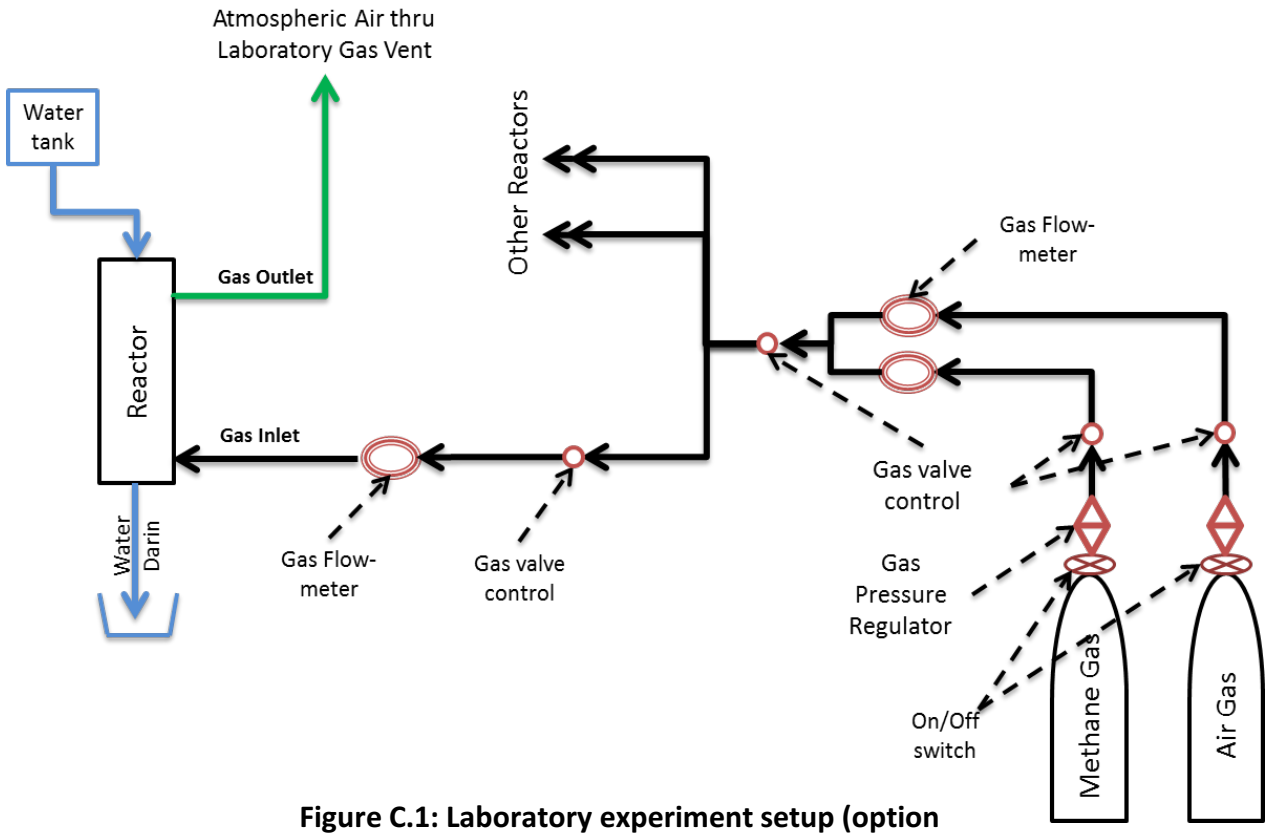


Figure C.1: Laboratory experiment setup (option 1)

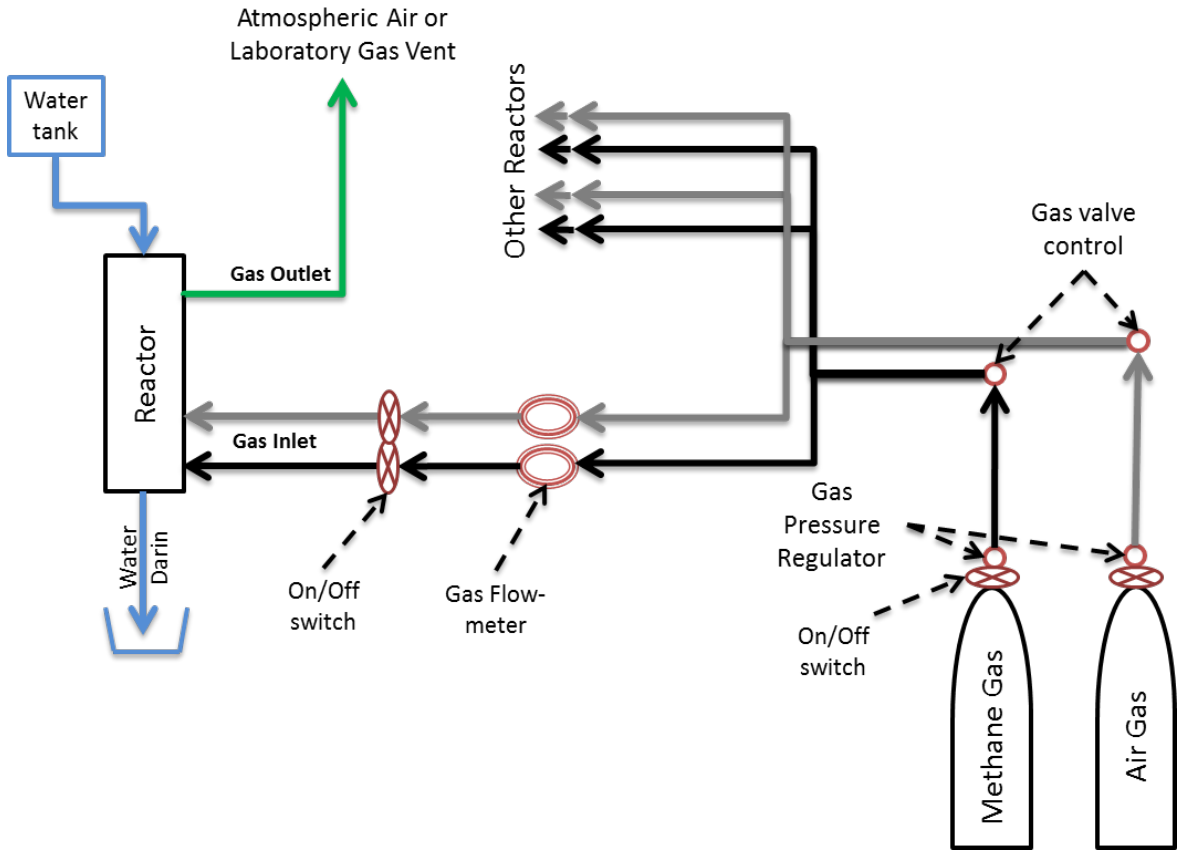


Figure C.2: Laboratory experiment setup (option 2)

Emergency measures

1- In case of any problems, turn off the gas cylinder from the main valve.
2- Contact project student or supervisor.

Scenario:

If it was a weekend and if all reactors leak all their methane all weekend, also if there was no room extraction (fans fail).

Laboratory volume: 6m x 20m x 3m x 1000 L/m^3 = 360,000 L

Maximum amount of Methane used thru the day (*if pure methane is used thru the system*):

10 ml/min x 60 min/h x 0.001 ml/L x 24 h/day. = 14.4L per reactor/ day

7 Reactors = 100.8 L methane per day

Amount of methane if all reactors leak their methane, all weekend with no room extraction:

So, 100.8 L methane per day x 2 days = 201.6 L /2 days

201.6 L in 360,000L if fully mixed = 0.056 % in Air (which is not flammable)

If poor mixing in room would need to have the 201.6 L methane contained in the local 4000 L of room space to reach 5% in Air,

i.e. all retained in local 1.8 m x 1.5 m x 1.5 m = 4.05 m^3 = 4050 L (VERY Unlikely)

In real situation the laboratory is well-vented by the air ducts and the air vent systems, also to add further safety factor, pure methane is not used with in the reactors, but a mix of 20% methane with air to add further safety factor by reducing the methane concentration.

Appendix D

Funnel Barrier System for Enhanced Oxidation

Funnel barrier system for enhanced oxidation

D.1 Introduction

The final objective of this study, as proposed at the outset of this investigation, was to understand all the main factors affecting methane production emitted from landfills before and after closure. In addition, the goal was to Identify potential engineered solutions for new/old landfills in developed and developing countries, which may enhance oxygen transfer in soils based in published literature and on the experimental results. In this chapter, an attempt is made to design a system based upon the information gained from the existing literature and the results from the experiments in the previous chapter, which can encourage an efficient interaction of oxygen with methane in the soil when compared to existing landfill cover mitigating methods. However, before identifying such systems, it is worthwhile to summarize and analyze the findings and relevant information gathered from the literature and experiments.

D.2 Summary of literature survey and experimental findings

It is imperative that the factors investigated and reported in the literature, as well as the results obtained from the experimental tests explored in Chapter IV, are taken into consideration when designing a cover system for the mitigation of landfill gases. If an optimum system is to be designed, all these factors and relevant details must not contradict, oppose, or have adverse effects on each other. However, not all of the factors that have been discussed in Sections E.1-E.24 (Appendix E) and shown in Table 2.9 are within the control of the designers. Additionally, the factors that could be managed and could be put at the disposal of the designing engineers and landfill owners are not all economically viable or practically feasible. For example, it would not be practical or economically feasible for landfill owners to cover the whole landfill site for a depth of 60-100cm with materials that are made of compost, and subsequently use this as a bio-cover system to mitigate methane. Moreover, municipal/regional/international laws and regulations may also limit the factors

to be available for control. Some regulations may stipulate that the cover layer must be made of low permeability materials, which is a property that would limit moisture and air from diffusing into the cover system (such as the EU regulation 1999c, Eureopean Directive 1999/31/EC, 1999). This All that leaves only a few factors that are manageable and controllable at the disposal of researchers, engineers, planners and owners to contend with, in order to obtain an optimum system. Some of the manageable factors that could highly influence oxidation are: oxygen availability in the soil and the type of soils supporting oxygen penetration throughout the depths.

The decision to place a certain cover on a landfill entails choosing controllable soil characteristic factors that are essential for the methanotrophic bacteria to function properly. Characteristic factors, such as soil density, porosity, water retention capacity, hydraulic conductivity, diffusivity, permeability, and heat capacity are the factors needed for maximum oxidation of methane (Chapter II). However, the optimum of one element of the soil characteristics might not be good enough for the choice of an optimum of another element of the same soil. Example, fine-grained soils allow more specific-surface areas for methanotrophs to exist in larger numbers, which is in contradiction to the need of having a highly porous soil for oxygen diffusion. That is because, fine-grained soils allow rapid clogging of soil pores due to the characteristic of high water retention capacity, hence preventing oxygen from diffusing to within these pores. Having combined optimum values of all of these factors specified in Table 2.9 for soil characteristics would consequently lead to optimum cover characteristic and hence, optimum landfill covers. Unfortunately, the type of soil that has these combined optimum characteristics does not exist. While a large number of materials and their combinations had been tried at one time or another (Huber and Lechner, 1999), thus far, no researcher has ventured on suggesting a specific soil type for a cover layer with such optimum characteristics. Nevertheless, Scheutz et al. (2009b) provided general guidelines that specified characteristics for cover layers as follows:

- A good oxidation layer should possess a long-term nutrient supply of nitrogen (N) and phosphorous (P).
- A layer should have high- temperature-insulating capacity.

- A layer should possess physical properties that could provide good porosity (for oxygen penetration) and good gas permeability, even at high water holding capacities.
- Compost materials when used as cover layers should have no artificial compaction.

Although these characteristics of a material are difficult to find in a specific soil material; a widely used material to oxidize methane is compost, proven to have efficient performance in laboratory experiments, with oxidation rate reaching up to 100% (Figure A.2). Huber-Humer et al. (2008, 2009) and Scheutz et al. (2009b) stated that, in order to reach the high rate of oxidation, the composted material must have the characteristics as follows:

- Well-matured, with high quality porosity.
- Well -structured substance, rich and stable in organic matter.
- High water holding capacity.
- Appropriate nutrient level.
- Permeability and fine texture to allow retention time for reactions.
- Sufficient volume of air-filled pores for gas flow.

The problem however, is that all these characteristics in a compost material are difficult to find in practice, alongside with the insurmountable disadvantages, as discussed in Appendix E and section 2.6. One particular disadvantage is that it does not satisfy the regulations prescribed by the European Commission (EC) 1999 (Eureopean Directive 1999/31/EC, 1999), in addition to other set regional laws and regulations. These regulations are meant to prevent the use of highly porous covers for landfills, which allows less methane to escape, as well as preventing the formation of leachates, in addition to avoiding the other technical difficulties and utilizing composted materials as a cover or part of a cover.

The other essential factor investigated in Chapter III was the availability of oxygen in the soil. Conclusively, investigations have not only emphasized the importance of oxygen availability to produce proper methane oxidation, but more importantly, they have the sustainability and continuity of oxygen supply inside the soil, therefore benefiting the methanotrophic bacteria with their needed oxygen.

The supply of oxygen into the oxidation layers comes from two sources. One comes from the ample air trapped in voids and cracks formed during the landfill cover construction, whereas the other source comes from the diffusion of atmospheric air into the soil through the surface. The first source, as explained in Appendix E, is unsustainable, but the second source is diffusion-dependent. Diffusion of air into the soil depends on the fluctuating atmospheric pressure, alternating wind speeds as pumping mechanism, as well as the soil characteristics of porosity and grains/voids structure inside the soil as factors affecting diffusion. In addition (but with much less influence) a third source of oxygen is a molar diffusion of air through the surface of the soils, which only penetrates the very top of the surface of the landfill cover, as discussed in Section E.9 (Appendix E). These mechanisms of supplying oxygen to the inside of the soil cover can usually penetrate only 5-15cm depth from the surface (Section E.3, Appendix E). This has been confirmed by several studies; one of which was reported by Huber-Humer (2004) and a second by Scheutz et al., (2009b) (Figure E.6, Appendix E). The importance of transporting oxygen into the soil in an adequate and sustainable level stems from the fact that not only is the methanotrophic bacteria present but there also other microorganisms in the same space of the soil, each competing for its own share of air. These other existing bacteria are as important as the methanotrophs, since they also have functions of their own; some can oxidize other toxic gases, such as the hydrochlorofluorocarbons, remediating the soil, while others provide nutrition to the vegetation soil, etc. In light of this, it has been found that oxygen consumption by other organisms can reduce methane oxidation substantially if the supply of air is not enough for all (Kettunen et al., 2006).

The soil characteristics, particularly the porosity of the soil, can affect the supply of oxygen and the degree of penetration into the oxidation layer. To show this mechanism and the oxygen penetration depth on methane oxidation, a lab column test was conducted by Molins et al. (2008) to study gas transport and degradation processes. They showed that the supply of oxygen and its degree of penetration could actually affect methane oxidation, Section E.12 (Appendix E).

"Many of the key factors for good performance have been identified. Nevertheless, there is still much work to be done to translate these findings into technical design and performance assessment guidelines that will ensure good methane removal, but allow for continued innovation and cost reductions." This was the conclusion reached by Huber-Humer et al., (2008), even with some advancement of landfill cover technologies at that time, (patented system by Ettala and Vaisanen, 2001, Figure A.3). All of these technologies that concern landfills were the technologies introduced and discussed in Appendix A. Bio-covers, bio-windows, biofilters, biotarps, and bioactive intercepting trenches, are all design concepts that were intended for the reduction of methane gas emission, and have advanced little with minor variations on the original concepts. The most concern amongst researchers regarding these design concepts is related to the cover material to enhance oxidations (Bender and Conrad, 1995; Boecks et al., 1996, Streese et al., 2001; Streese and Stegmann, 2003; Hilger and Humer, 2003, Schuetz and Kjeldsen, 2004; Humer and Lechner, 2005, Haubrichs and Widman, 2006; Enola et al., 2007; Philopoulos, et al., 2008, Einola, et al., 2009; Ren, et al., 2012; Reddy, et al., 2014; and some others), particularly given the large number of parameters involved with cover materials. However, a small amount of research had been done on a meaningful technological advancement to a next level. Unfortunately, most of these efforts concerning the research of bio-cover materials and research on the parameters affecting oxidations have been conducted for the purpose of finding an ideal material that can be structured for optimum oxidation, in which actual materials could not be feasible in real practice; thus, rendering further efforts in this direction, ineffective.

Although cover concepts were designed successfully and have been shown to produce some enhancement for methane oxidations, each design concept has not been without its own drawbacks. Total bio-cover system, open bed biofilters, bio-windows and bio-tarps are all concept designs that have bed surfaces open to the atmosphere. That in itself is in conflict with regulations, since it could allow methane to escape freely through the surface, and allow rainwater to seep down through these open bed systems, creating leachates polluting the underground water. Aside from compliance issues with regulation, other difficulties with these systems are the cost of building, transportation and distribution of materials over the entire surface of the landfills, as well as to the costs of the materials themselves. In addition,

the availability of materials suggested by these concepts, which require certain specific characteristics, i.e., sizes of aggregates, organic compositions, construction materials and construction equipment,…etc. which may not be available locally or may not even be available for some countries or regions. Some concepts require active collection of landfill gases via pumping systems, such as the active open/closed bed biofiltration systems, which then utilize the gases into biologically active biofilters for methane oxidation. In addition, other concepts may require an aeration of the biofilters using an active pumping, which requires costly operation and maintenance. These drawbacks have stood as obstacles to municipal localities and/or institutions to accept these concepts for landfill gas control.

Methanotrophs serve as the working engine for landfill gas removal, whereby methane oxidation by methanotrophs occurs in soil layers for both in high and low levels of oxygen concentrations. Type I methanotrophs have a membrane-bound particulate methane monooxygenase enzyme (pMMO) and can perform more efficiently under low methane, high oxygen environments. In contrast, Type II methanotrophs use their soluble methane monooxygenase enzyme (sMMO) in the presence of copper and perform more efficiently under a high methane, low oxygen concentration environment. Thus, the appropriate environment for Type I is when oxygen is at the concentration level of 21% vol/vol; while 1% vol/vol is ideal for Type II. The significance of this is that atmospheric oxygen concentration would yield dominant methanotrophs of Type I, due to the high oxygen concentration in the upper soil, while Type II would be present in the lower levels of the cover layers. These methanotrophic bacteria have been researched extensively within literature (Whittenbury et al, 1970; Anthony, 1982; Gommers et al., 1988; Mancinelli, 1995; Hanson and Hanson, 1996; Dalton, 2005; Watzinger et al, 2006; Dunfield et al, 2007; Conrad, 2009; Jugnia et al., 2009; Scheutz, et al., 2009b); their genera have been identified and their properties catalogued (Table 2.1). Furthermore, the capacity of methanotrophs to mitigate methane under various concentrations can only reach maximum levels under the conditions discussed in Appendix E, which is a reality that must be dealt with when designing an optimum cover.

Methanotrophs require oxygen so that they can actively convert methane into carbon dioxide and other substances according to equations 2.1 and 2.2, such as the following:

Ribulose monophosphate pathway (RuMP):

$CH_4 + 1.5\ O_2 + 0.118\ NH_4^+ \rightarrow 0.118(C_4H_8O_2N) + 0.529\ CO_2 + 1.71\ H_2O + 0.118\ H^+$ (D.1)

The Serine pathway:

$CH_4 + 1.57\ O_2 + 0.102\ NH_4^+ \rightarrow 0.102(C_4H_8O_2N) + 0.593\ CO_2 + 1.57\ H_2O + 0.102\ H^+$ (D.2)

These equations would imply that methanotrophic bacteria require three moles of oxygen for each of two moles of methane in the Ribulose pathway process, and little more than that for the Serine pathways. The significance of this is that the oxygen supply from the atmosphere may not be enough to support the activity of this bacterium, given the fact that methanotrophic bacteria and other microorganisms present in the soil are in direct competition for the same resource. This dire demand for oxygen supply by these microorganisms in the soil has since been well recognized by researchers and has resonated in their citations as follows:

- "Most methanotrophic bacteria are obligating methanotrophs and strict aerobes" (Hanson and Hanson, 1996). "Their activity depends on the presence of sufficient concentrations of both CH_4 and O_2; and thus, they tend to be confined to fairly narrow horizontal bands within their habitat, limited in their distribution by the downward diffusion of atmospheric O_2, and the upward diffusion of CH_4" (Scheutz et al., 2009b).
- "To the extent that O_2 availability limits oxidation, cover designs that enhance O_2 availability will enhance CH_4 oxidation" (Barlaz et al.,2004).
- "High methane degradation rates can only be accomplished if the supply of atmospheric oxygen to the methanotrophic community is adequate (Gebert and Groengroeft, 2009).

It is unfortunate that no advancements have yet been achieved concerning oxygen supply and its availability. The only supply of oxygen available for all these design concepts presented by researchers are the ones that rely on atmospheric diffusion due to molar and

barometric pressures. However, the supply of these sources in themselves are unpredictable and are inefficient in their way of delivering oxygen, particularly if the atmospheric pressure is equalized or is run up by advective upward gases from the landfills. Therefore, if a system could supply air in an adequate and controlled manner, whilst also avoiding the majority of the drawbacks associated with current available design concepts, with little or no added costs as of active systems or elaborate builds, this would unquestionably be an added advancement to the accomplishments in this important area of landfill gas control and mitigation.

The experimental investigation in Chapter IV revealed that oxygen availability within the bacterial community in the soil is essential, illustrating its profound effects on the oxidation of methane. It was shown that when oxygen was made available to the methanotrophic bacteria in a multitude of samples, regardless of the type of soil or the level of exposure to methane previously, the bacteria responded well and oxidized methane in the same manner in all of the samples tested. The bacteria consumed methane aggressively and continued this course of consumption linearly, reaching close to complete consumption. This clearly meant that if an oxygen supply were to be made constantly available, the methanotrophic bacteria could consume methane aggressively in the same aggressive linear relationship regardless of other factors. This important information is critical for engineers to be aware of in order to design and construct an effective methane reduction cover.

The other experiments, performed through column reactors, also showed that common and plain soil, mixed with sand and having no added organic matter as a landfill cover, could not support oxidation well. The significance of this is that these widely available materials, particularly in arid zones, could not be relied upon for methane mitigation, and therefore an alternative and effective system must be designed for such environments. Finally, it was shown through a continues flow reactor experiment (CFR), that when oxygen was made available deeper within the layer of the soil and close to the methanotrophic bacteria community, a much higher rate of oxidation was achievable, reaching up to 65% higher compared to when oxygen was only made available at the surface of the soil.

Taking into account the discussion within this section, and the information gained from the continuous flow reactor experiment (CFR), investigating the effect of supplying oxygen deep within the soil (Section 4.3), which showed profound effect on the oxidation efficiency, an attempt was made to venture into proposing a new innovative gas mitigation system described in the following sections.

D.3 System description

Based on the arguments presented in Section D.2, the importance of supplying adequate and continuous oxygen to the living microorganisms in the landfill soil for their essential needs of oxygen and nitrogen was a key component for the system design. A system that utilizes the pressure of wind to channel air into the deeper layers of cover soils has therefore been proposed. This innovative system consists of a wind gathering concentrating cone, a one-way gate valve system and a delivery pipe system (Figures D.1 and D.2). The whole concept depends on utilizing the wind energy to be the passive driving force for oxygen into the soil layers, with an important gate system to prevent an inverse of gas ventilation from moving up and out of the landfill layers. This gate system would allow a one-way passage of air in, and also prevent gas from coming out. Furthermore, it is important that this is installed to also constrict methane and other hazardous gases from leaving the ground layers through the less resistive passage of the pipe. Thus, it shall be designed only for the passage of air to the areas of methanotrophic communities, as well as a means of preventing atmospheric pollution. Additionally, at the lower end of the pipe system, a perforated section will be included to ensure a maximum dispersion of air to reach a wider soil surface area.

As illustrated in Figures D.1 and D.2, this design has purposefully been kept very simple, employing a low level of technology, and constructed with limited parts and limited amount of materials such as simple metal sheets and pipes or potentially even from discarded materials like plywood, plastics or discarded pipes. The reason for this is that this would be suitable for implementation in any country or region, particularly in those that are developing. A prototype of this system had been build and operationally tested for its functionality.

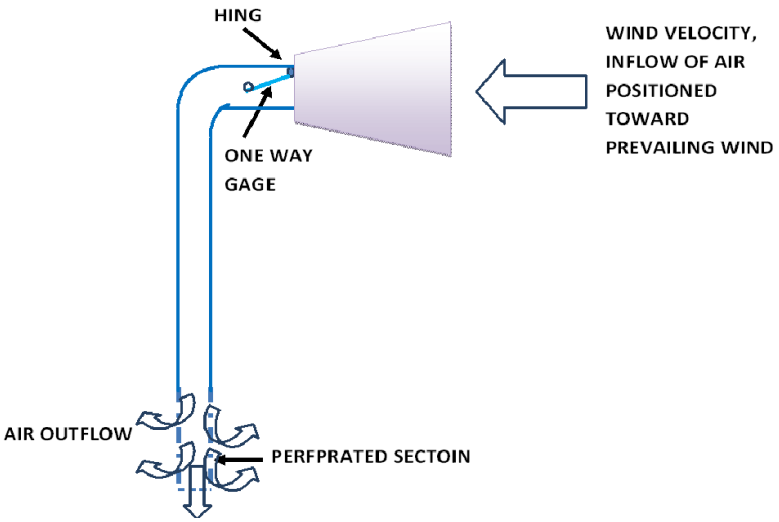


Figure D.1: Schematic diagram of the system's configuration.

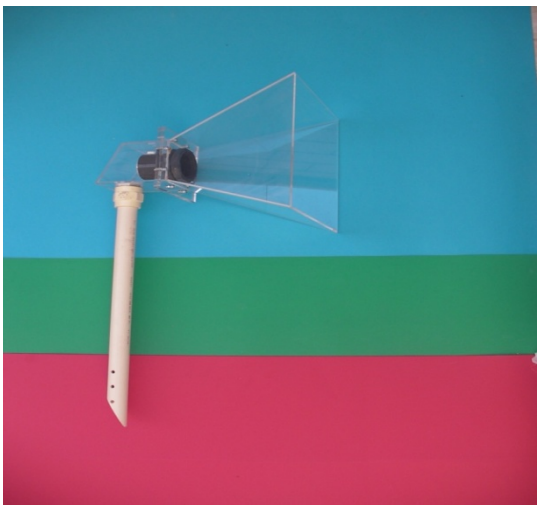


Figure D.2: Image of wind- forced diffusion system.

D.4 Systems operations

Air pressure enters from the wider end of the cone and gathers more pressure due to the gradual narrowing of the cone at the narrower end, pressuring the gate valve to open, whilst continually moving with its higher pressure down to the oxidation layer (as indicated in Figure D.3). Due to this higher air pressure from above, air is distributed through the pores and cracks within the oxidation layers, reaching and nourishing the microorganisms of the soil, and allowing the oxidation process of methane to take place more efficiently before reaching the upper layers. The result of oxidation is mostly carbon dioxide and water, which

will then slowly disperse and leave the surface of the landfill, as shown in the schematic diagram in Figure D.3. In case the prevailing pressure at the inflow of the concentration cone becomes low, the higher gas pressure inside the landfill layers and the delivery pipe will force the gate valve to shut during this high landfill pressure, as indicated in Figure D.4. The gate valve was designed to close at any instance of higher or equal pressure inside the delivery pipe. Figure D.4 shows the gate valve in the shut position and in an open position when air is blown directly to the system by an air blower.

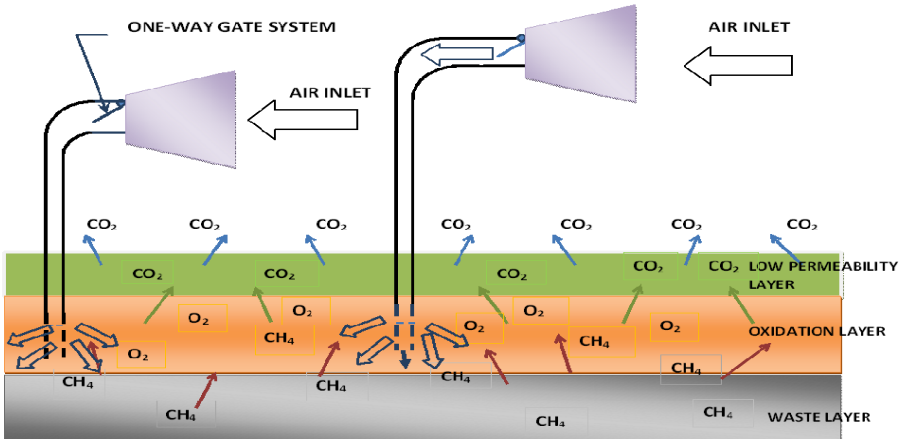
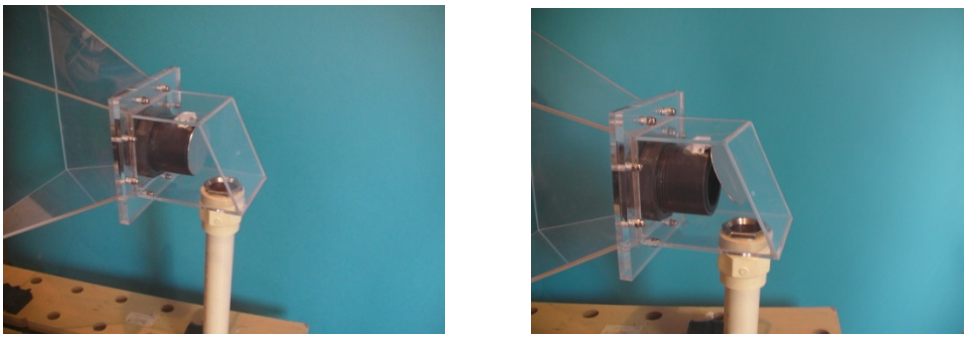


Figure D.3: Schematic diagram of the system's operation in a landfill, using multitude of devices.



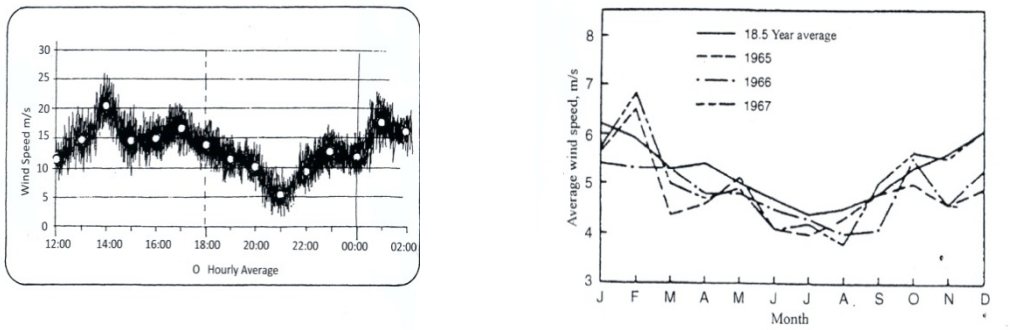
(a) (b)

Figure D.4: The gate valve positions (a) closed and (b) open, due to induced air pressure.

It is important to note that the system's dimensions are site-dependent. That is, the wind concentrating cone diameters, lengths, directions, and the pipe system heights can be varied and adjustable depending on the landfill site. For low wind blowing areas, a wider cone for gathering more air pressure is constructed, along with higher elevation of the pipe system and *vice versa*. The system could also be designed so that the cone part can follow the direction of the blowing wind in an automatic fashion.

Wind power and direction are important factors in the design of this system, and data for both of these can be readily available from local meteorological stations. For example, the average wind pattern of half-day wind speed for the United Kingdom is shown in Figure D.5a, and seasonal average wind variation for the eastern United States is shown in Figure D.5b (Manwell et al., 2009; Twidell, 1987). Wind with a speed of 5 m/s could produce a power per unit area equal to 80 Watt m^{-2} (Manwell et al., 2009), which is suitable for passing the appropriate amount of air into the ground soil. Both of these figures serve as demonstration for wind and direction patterns, which could differ from one region to the other; that said, the direction of wind however, predominantly followed the worldwide wind circulation pattern, as shown in Figure D.6.

The landfill cover technologies that have been discussed in Appendix A were seen to be dependent on having a methane distribution layer constructed throughout the landfill site and made of coarse material for the purpose of distributing methane gas uniformly before the gas could reach the oxidation layer. This was necessary because the oxidation layer had very shallow oxygen penetration. However, in this new concept, there is no need for such a costly layer to be constructed, since the oxygen air is readily delivered and distributed at any desired depths and locations within the landfill. Therefore, this implies saving in the costs of construction, and would increase oxidation by the methanotrophic bacteria, deep down in the soil before these gases could reach the top cover layer. While added coarse material around the perforated section of the delivery pipe inside the layers could be an added advantage to the distribution mechanisms, it is unnecessary in the presence of high-pressure gradient.



(a) (b)

Figure D.5: (a) Wind speed average for half a day, U K (source: Twidell, 1987), and (b) monthly wind speed average over a year time frame, Montan Billings, USA (source: Manwell et al., 2009).

Placement of this new system on surfaces of landfills can be fairly flexible. A multitude of systems can be planted all over the surface of the site, spaced equally or randomly, facing known prevailing wind directions or placed in different directions to catch wind pressure from all directions, as illustrated in Figure D.3. The system can also be installed as one central system, in which only one larger configuration is embedded in the site. The multitude system configuration can be installed in the same frequency similar to the installation of biofilters, or bio-windows systems, while the central system can be installed alone, but could involve added distribution and ventilation piping, as illustrated in Figure D.7. Although this could involve an addition to the initial capital costs, installing such a system could prove to be more aesthetic.

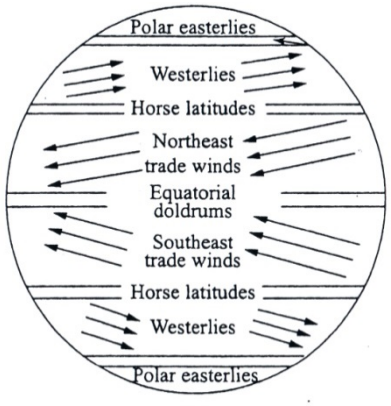


Figure D.6: Prevailing global wind patterns (source: Manwell et al., 2009).

It is worthy to note that this new concept of air delivery system is direct, passive, and operation free, requiring little or no maintenance, thus less cost. The concept gives complete air supply control to the landfill engineers for their assessment of the amount of pressure and air quantities that needs to be transported inside the layers of a particular landfill, which is contrary to the diffusion-dependent concepts. More importantly, the delivery of the pipe system could be driven directly into the landfill surface without disturbing the surfaces, particularly for old landfills. Moreover, instead of the biotarp system, this system could be installed directly over the intermediate covers during the active landfilling, whereby it could be placed and removed readily without surface disturbances. Furthermore, the concept could work well with completely sealed landfills containing hazardous substances, provided that a proper ventilation system could be installed alongside it.

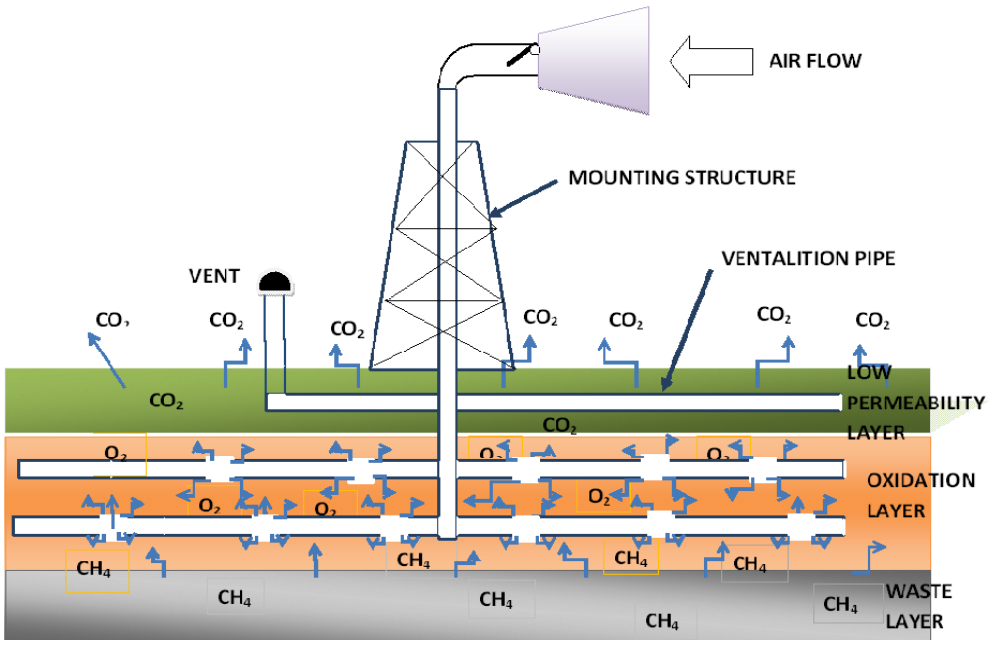


Figure D.7: Schematic diagram of a central system concept.

In applying this new design for Kuwait, the country is an arid region that receives no more than 10cm of rainwater in a year. Thus, an added advantage of this system, particularly for

this country and some other arid countries, is that the concentrating cone part of the system can be tilted a little upwards to catch some rainwater, which can to be transported down with the air into the landfill layers, and in turn, provides moisture on its way down inside the soil.

D.5 Pressure build-up estimation

When wind is blowing in a speed of V_1 into the open mouth of the system's concentrating cone (Figure D.8), it creates a pressure front that will travel throughout the system, reaching down to the inner layers of the landfill soil. Designing and constructing appropriate parts of this system require some knowledge of the amount of pressure and the quantity of the air that could be created from the blowing winds, in order to ascertain and select the appropriate cone size for a particular landfill site job. To calculate the pressures developed at both ends of the system produced by the wind energy, as it is created by the differential atmospheric pressures, this must be adhered to and accounted for. Thus, the kinetic energy of a mass of air Δm equal to the mass of the volume of air enclosed by the concentrating cone boundary, and the two sections 1 and 2 as a control volume (Figure D.8) when it is initially at the inflow section 1, the kinetic energy is given in the following equation:

$$KE_1 = \tfrac{1}{2}\,\Delta m\, V_1^2 \qquad (D.3)$$

When the same Δm reaches to section 2 from section 1 and after sometime (Δt), the increase of the kinetic energy at the second, section 2 is also given in the following equation:

$$KE_2 = \tfrac{1}{2}\,\Delta m\, V_2^2 \qquad (D.4)$$

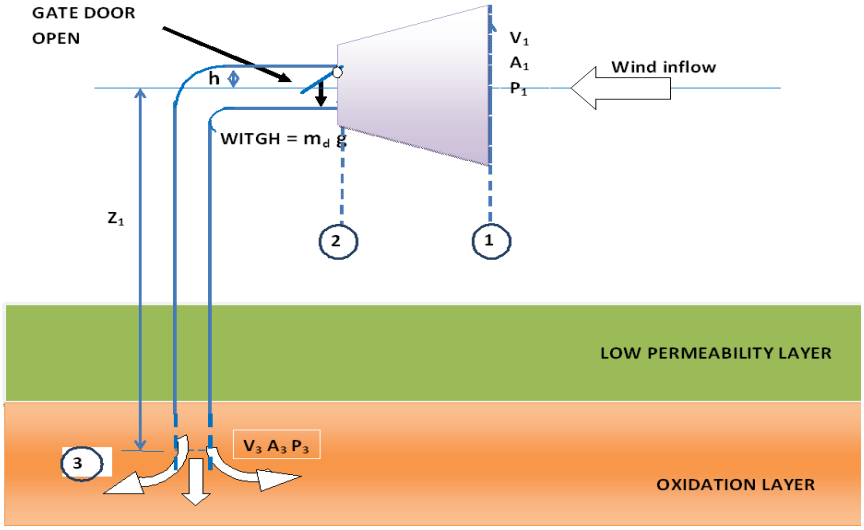


Figure D.8: System controlling variables.

Along the path of the pressure front created by the wind speed, some friction could be encountered, however this friction energy is assumed negligible. Furthermore, the flow of air will be assumed continuous and incompressible. This latter assumption is accurate for fluids and correct for gases if the flow of air velocity speeds does not reach a Mack number (Mack number of 304.3 m/s), it will produce negligible errors. There would likely be errors less than and up to 5% for speed of up to 213m/s (Noel de Nevers, 2005). In addition, it will be assumed that the system does not create heat or lose heat (i.e., a conservative system). The pressure force p_1 at the inflow of the system will also do work, as the mass element Δm initially at section 1 will be moved by a velocity V_1 to reach section 2 after some time Δt. Its kinematic movement will reach a distance d_1 after some time Δt, according to the kinematic formula,

$$d_1 = V_1 \Delta t \qquad (D.5)$$

Similarly, Δm at section 2 will move away, covering a kinematic distance d_2 according to the same formula as,

$$d_2 = V_2 \Delta t \qquad (D.6)$$

Equations D.5 and D.6 represent displaced masses of air at the inflow and outflow of these two sections, and if each of the equations were to be multiplied by the density of air (ρ) for their corresponding sectional areas, the result would be equal to the mass of the displaced fluid, according to the continuity of fluids, as follows:

$$\rho A_1 d_1 = \rho A_2 d_2 = \Delta m \qquad (D.7)$$

Using equations D.5 and D.6 into equation D.7, the result would be the following:

$$\rho A_1 V_1 \Delta t = \rho A_2 V_2 \Delta t = \Delta m \qquad (D.8)$$

By definition, work energy of a force is equal to the magnitude of that force multiplied by the distance it travels. Therefore, the pressure at section 1, p_1 causes a force equal to $p_1 A_1$, and similarly, the force at section 2 is equal to $p_2 A_2$; hence, the work done by each force is given by the following:

$$W_1 = \text{Force x distance at section 1} = p_1 A_1 d_1 \qquad (D.9)$$

$$W_2 = \text{Force x distance at section 2} = p_2 A_2 d_2 \qquad (D.10)$$

Using equation D.7 to substitute for d_1 and d_2 in equations D.9 and D.10, the following work energy would result to the following:

$$W_1 = \Delta m (p_1 / \rho) \qquad (D.11)$$

$$W_2 = \Delta m (p_2 / \rho) \qquad (D.12)$$

Energy will be gained as a potential energy, due to the work required to lift up the gate door valve at section 2. This potential energy would amount to the following:

$$PE_g = m_d g h, \qquad (D.13)$$

Where, m_d is the gate door mass, g is the gravitational acceleration and h is the height of the center of gravity of the gate door that will be moved up due to the pressure from the flow.

Employing the conservation of energy principle, which accounts for all energies made or lost in the process at both sections, would result in the following:

$$\sum KE_1 + \sum W_1 + \sum PE_1 = \sum KE_2 + \sum W_2 + \sum PE_2 \qquad (D.14)$$

Using equations D.3, D.4, D.9, D.10, and D.13 into D.14, and rearranging would yield the following:

$$½ V_1^2 + (p_1/ \rho) = ½ V_1^2 + (p_2/ \rho) + m_d gh/ \Delta m \qquad (D.15)$$

The last term of equation D.15 consists of the mass of the gate door and the height h that it will raise in response to the pressure of the flow. Both of these quantities are small in relation to the mass of the control volume Δm, but become much smaller when compared to the other terms of the equation that consisted of the pressures and the squared velocities of the flow. Therefore, this term can be ignored for this type of approximation; nonetheless, should there be a need to have better approximation, as then it could be included in the analysis with little evaluation works. With that assumption in place, the equation is reduced to the well-known Bernoulli's equation, evaluated for the flow of air between sections 1 and 2 of the system, as follows:

$$½ V_1^2 + (p_1/ \rho) = ½ V_2^2 + (p_2/ \rho) \qquad (D.16)$$

When rearranging for the pressure p_2, the equation is reduced to equation D.17.

$$p_2 = p_1 - \rho (V_2^2 - V_1^2)/2 \qquad (D.17)$$

The same procedure can be repeated at the part segment of the system, lying between sections 2 and 3 (Figure D.8), which would include the loss of potential work of the weight Δmg to move a height z_1 from position 2 to position 3, along with the gravitational pull. If all energies are to be accounted for, namely the kinetic energies of the air flow due to the velocities V_2 and V_3 at sections 2 and 3, then the pressure works done by p_2 and p_3 at the two sections and the potential energy done by the weight Δm g, and again by employing the principle of energy conservation, another Bernoulli's equation for air movement from section 2 to section 3, as in the following: (return to the original)

$$\Delta m\ V_2^2/2 + \Delta m\ (p_2/\rho) + 0 = \Delta m\ V_3^2/2 + \Delta m\ (p_3/\rho) - \Delta m g z_1 \qquad (D.18)$$

Solving for p_3,

$$p_3 = \rho\ (V_2^2 - V_3^2)/2 + p_2 + \rho g z_1 \qquad (D.19)$$

Substituting equation D.17 into D.19, and rearranging it would produce the following:

$$p_3 = \rho g z_1 - \rho\ (V_3^2 - V_1^2)/2 + p_1 \qquad (D.20)$$

This last equation describes the relationship between the pressure at the end of the delivery pipe of the system in Figure D.8, in terms of the velocities at the inflow and outflow of the system, with including the height of the system above ground up to the end of the delivery pipe. However these relationships could be difficult to measure; thus, if the equation could be expressed in terms of the sectional areas and the inflow velocity, then the equation could be more practical and useful for landfill site engineers. Employing therefore, the continuity principle for fluids, which states that the mass rates at different points of fluid flowing through a container are equal (i.e. if the fluid is incompressible, in a steady state, and is conservative (no loss or gain of heat)), the equation of continuity would be as follows:

$$\rho A_1 V_1 = \rho A_3 V_3 \qquad (D.21)$$

Rearranging for V_3, gives the relationship:

$$V_3 = V_1 \ (A_1/A_3) \qquad (D.22)$$

Substituting equation D.22 into D.20 and rearranging for the pressure drop Δp would give the following:

$$\Delta p = \rho V_1^2 \ (A_1^2/A_3^2 \ - \ 1)/2 \ - \ \rho g z_1 \qquad (D.23)$$

Equation D.23 gives the pressure drop across the system, starting from the inflow of air at section 1 through the system, ending at the diffusion point of the delivery pipe at section 3 (as shown in Figure D.8), in terms of the inflow velocity V_1 and the relative cross-sectional areas of the inflow and the outflow sections A_1 and A_3. The equation could also be used to determine the required pressure inside the soil, p_3, in order to construct a system that produces a desired air diffusion rate that is suitable for a particular soil of a landfill. In equation D.23, the standard air density (ρ) is 1.225 Kg/m^3 (0.0765 lb_m/ft^3at sea level at 15° C). The gravitational acceleration (g) is 9.81 m/s^2 (32.2 ft/s^2); while the pressures p_1 and p_2 are the absolute pressures, given in terms of the gauge pressure and the atmospheric pressure, as in the following (Noel de Nevers, 2005):

$$P_{abs} = p_{gage} \ + \ p_{atm} \qquad (D.24)$$

In addition to the aforementioned analysis, defining airflow equations, air volumetric flow rate inside the soil layers might also be an important factor that could be considered when constructing a system. By definition, the volumetric flow rate quantity for steady state and incompressible fluids is given by the following:

$$Q = V_1 A_1 = V_3 A_3 \qquad (D.25)$$

This equation is useful in determining the anticipated amount of air to be delivered to the inside of the soil layers.

It could be observed from the air diffusion expression of equation D.23, that the diffusion mechanism is clearer and the air diffusion amount is easier to measure. This is contrary to the diffusion mechanism incurred due to the wind and the barometric pressures used to find the rates of diffusion of oxygen for soils in the systems of concepts discussed in Appendix A. It should also be noticed that the diffusion of air into the soil for both of this concept in Figure D.8, and those of the other designs in shown in Appendix A, that all of these systems depend on wind and barometric pressures to deliver oxygen to the inside of the soils. The only difference between them is the type of delivery mechanisms used. The system described in this chapter relies on direct utilization of wind power to force air passively into the soil; while the other concepts depended only on the natural molar and surface pressure gradients to diffuse oxygen into the soil.

D.6 Anticipated performance

The new developed system of methane mitigation opens a new window, and possibly, a new direction in research, and likewise opens horizons toward proposing a new oxygen delivery system and other anticipated similar systems. This new concept has never been tried before in the mitigation of landfill gases, thus the theoretical foundation for this new methane reduction system is in need of experimental verification. However, the set of factors that will influence performance of the system must first be recognised.

The new system of delivering oxygen into landfill's soils, as previously described, has many flexible parameters that can be calculated in order to construct an efficient system that is suitable for a particular landfill site. From equations D.23 and D.25, the principal parameters for the system are, the inflow of wind speed, the cross- sectional areas of the wide end of the wind-gathering cone, the outflow wind speed at the end of the immersed delivery pipe, and the height of the cone above ground to the end of the delivery pipe. This group of factors is related to the system's design and construction, summarized as follows:

- Inflow velocity, V1.

- Cross-sectional area of the wide end of the concentration cone, A_1.
- Cross-sectional area of the end tail of the delivery pipe, A_3.
- Height of the system above ground down to the end of the embedded delivery pipe, z_1.
- The gauge pressure at the inflow of air, p_1.

This group of factors is area, country, and regional dependent; therefore, in order to verify their performance, a field test involving another research venue and time allocation must be undertaken.

The other group of factors that affect oxidation performance are the ones that deal with the landfill location and characteristics of the ground, waste, environmental, meteorological, and finally, soil and cover. These groups of factors are listed in Table 2.8. It is important to note that adequate research has been done to verify the effects of each one of these factors on the oxidation process, as discussed in Section E.1 through E.24 (Appendix E). However, these numerous factors were all done on conventional and top cover material-based techniques, as sown in Appendix A. These numerous factors must now be evaluated for this new system. Although the anticipated performance of this new system can be observed from the experiment performed by Streese and Stegmann (2003) on an active closed bed biofiltration system (as discussed in Appendix A), in which a high rate of oxidation was observed, reaching up to 1448- g CH_4 $m^{-3}d^{-1}$ when the system was supplied with oxygen via a pump (Figure A.7). In addition to this observation, in a municipal solid waste-simulated landfill experiment in a laboratory using lysimeter containers, Slezak et al. (2015) showed that even a little aeration rate ($4.41x10^{-3}$ L $min^{-1}Kg^{-1}$) introduced in the waste layer, can produce a high rate of carbon release due to oxidation (as much as five times more), when compared to the unaerated system. This indicates that higher rates of oxidation can be achieved efficiently if the supply of oxygen were to be made available and appropriately via some delivery systems, other than the passive systems supplied with oxygen via the natural atmospheric diffusion delivery mechanisms, in which rates of oxidation did not exceed more than 500-g CH_4 $m^{-3}d^{-1}$ (Table 2.6). Therefore, the performance of this new passive system with induced wind and atmospheric pressures as the delivery mechanisms is anticipated to perform as well, without

the drawbacks associated with the other systems. The anticipated performance could at least reach 65% performance over the natural air supply systems, which was the level of performance obtained from the CFR experiment, and could even reach higher percent rate if the delivery pipe were to be place deeper inside the soil. To arrive at an actual performance level for this new concept, a field test experiment that takes all of the environmental factors of Table 2.8 into consideration would be required, particularly the soil and cover characteristic factors (porosity, temperature, water holding capacity, permeability, acidity, nutrients and minerals, and inhibiting substances). Although this is a huge undertaking having to require extended research effort and time; however, this would open wider avenues for research on this new concept.

Laboratory experimentation with this new system is quite difficult, requiring time and cost investments. The testing would require special equipment to simulate wind fluctuations and pressure variations at the open end of the cone, and other special equipment to measure the wind and pressure outcome at the end of the immersed delivery pipe. Once the equipment are acquired, a design of the testing and ground soil formation must be fabricated, tested, and then contained from external elements for the actual testing. Additional equipment is also needed to test for each of the factors affecting methanotrophic behaviour and the oxidation levels. For example, this entails the need for devices for all and each of the factors affecting methane oxidation, with each factor requiring a different experiment setup and equipment. These sets of tests may take years, and therefore investments similar to the course taken to develop the other techniques and capture methods discussed in Appendix A. However, and more importantly, the safety and risks to facilities and personnel when using flammable and explosive mixes of methane and air, particularly when the experiments are left running unattended, is a very important factor for any institution to consider. In particular, if landfill simulation is required, where a high rate reaching up to 5 cm^3 min^{-1} of methane load is to be used for such anticipated simulation. This is the methane production rate realized by Kightley et al. (1995). The option left is to test and monitor this system in an open field, using the actual landfill parameters for the tests. Using the grounds of a landfill for testing would require different procedures altogether.

Landfills in Kuwait use impermeable soils (hardpan calcareous soils, a sandy matrix with high silica content, and slight Gypsiferous), which is laid intermediately and as a final cover, allowing very little atmospheric air diffusion to penetrate, hence higher anaerobic reaction in the waste. Therefore, the performance of this new concept system can increase oxidation to higher magnitudes, and does not have to deliver high quantities of air inside the layers, as only small amounts are sufficient to start the process of releasing carbon in the form of carbon dioxide and other less harmful gases from the landfill. This is in conjunction with a recent experiment, showing that high rate of aeration produced 26.5% of the carbon contained in the waste. In addition, a small aeration produced higher rate of 31.8%, compared to the unaerated lysimeter system, which produced only 6.9% (Slezak et al., 2015). This latter case of the unaerated system is a case for treating landfills when using the concept methods discussed in Appendix A. Besides the anticipated benefit of producing high rate of oxidation, establishing a system of low cost and low maintenance, and with little disturbances to the surface of the landfill, while complying with set regulations, this new system will have more additional benefits. Some of these benefits are the faster stabilization of the waste load, decreased amount of leachate and pollutants in the leachates, and the reduction of odour (Erses et al., 2008; El-Fadel et al., 2013).

D.7 Computer simulation of the funnel model

To find out about the extent of which the air (and oxygen) can diffuse inside a porous medium using the above discussed funnel system, a computer simulation program was attempted. Taking this procedure was an important step toward paving the way for setting up experiments to verify the proposed funnel system. Therefore, ANSYS finite element simulation program was utilised (http//www.ansys.com/products/academic/ansys-student; free download). For this simulation to be of relevance, however, published data of an active landfill must then be used. The data of Kelso Waste Depot landfill located in Sydney, Australia, using a biowindow filter (Cell D containing composted municipal solid waste amended with 10% shredded wood) was incorporated in the model (Dever et al., 2007, Dever, 2009, Dever et al.,2011). Fortunately, abundance of tutorials is available on the

internet, explaining the use of this particular code, and showing the straight-forward input procedure of the data. Some of these tutorials are as follows:

https://www.youtube.com/watch?v=UVMXAT1NU0Q&list=PLd23hHm4FCRclfHw4-W_rGQX2hJCNnPPK;

https://www.youtube.com/watch?v=1pxvdwEWc5Q;

https://www.youtube.com/watch?v=qvjHeVc-i8E;

https://www.youtube.com/watch?v=N6_EiMra9cw;

https://www.youtube.com/watch?v=7cEI4bnV0_4;

https://www.youtube.com/watch?v=HiqQfr_8Ebw; .....

A cell of 3-m width by 1.2-m depth was used as a two dimensional porous model for the simulation, as indicated in Figure D.9, which duplicates the bio-window filter system that was placed on Kelso landfill. A mesh generating feature of the program was then utilised to produce triangular elements of the model, as shown in Figure D.10. The cell was also modelled, so that the outside atmospheric pressure and air data can be incorporated in the model as boundary conditions. The immersed pipe was also modelled as part of the boundary conditions, as shown in Figure D.11.

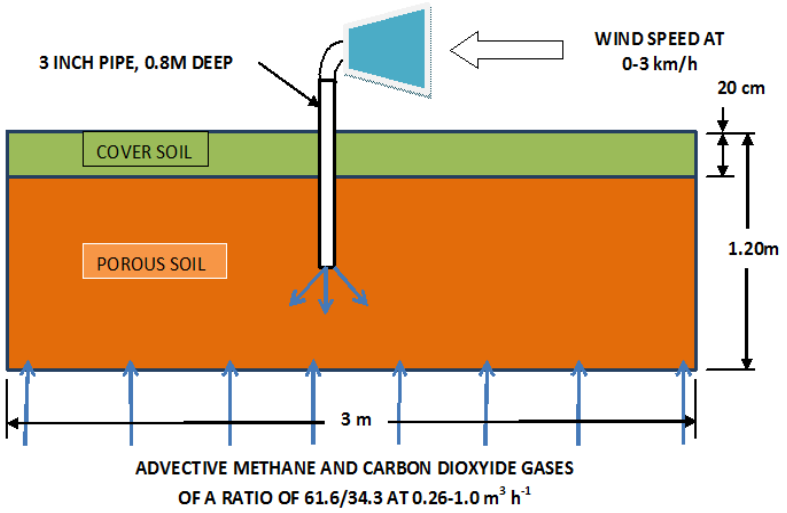


Figure D.9: Diagram of simulated model.

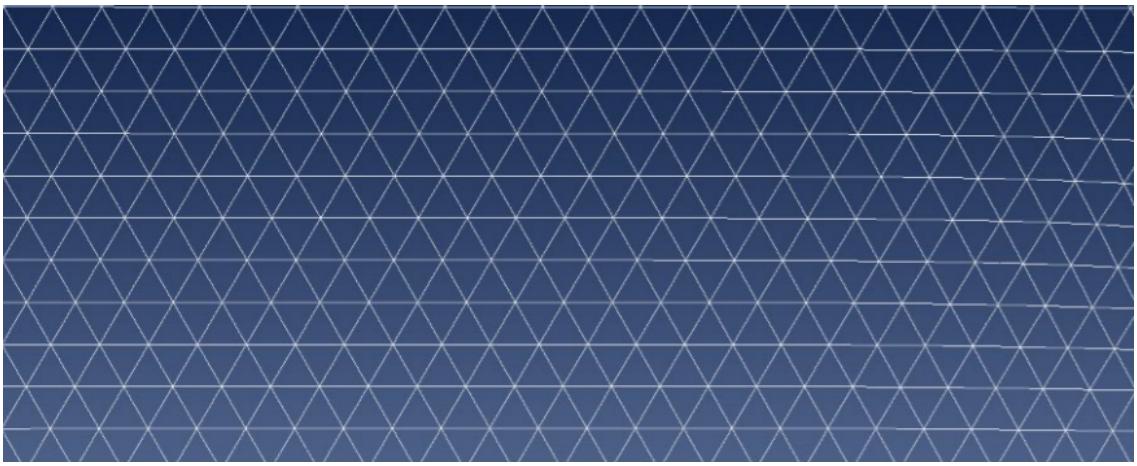


Figure D.10: Triangular elements used for the simulation.

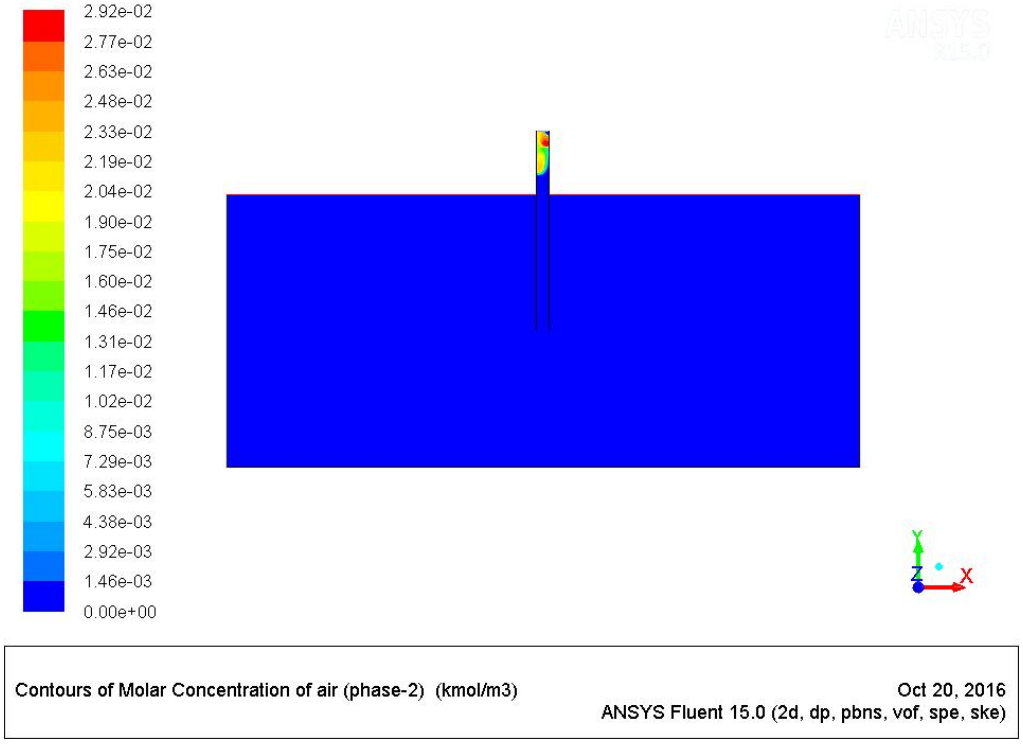


Figure D.11: Atmospheric air and immersed pipe as boundary condition of the model.

The data used for creating an input into the model was as follows:

- Wind speed= 3 km/h.
- Advective LFG gas rate at 1.0 m^3 h^{-1}.
- LFG composition 61.6:34.3, CH_4:CO_2.

- Atmospheric pressure 101.7 kPa.
- Pipe diameter 3 in.
- Pipe length inside the porous medium= 0.8 m.
- CH_4 viscosity= 11.01 Pascal.
- CH_4 density= 0.6335 kg/m^3.
- CO_2 viscosity= 14.72 Pascal.
- CO_2 density=1.743 kg/m^3.
- Air density= 1.225 kg/m^3.
- Hydraulic conductivity of the porous medium= 1.75 x10^{-5} m/s.
- Porosity= 0.495
- Outside temperature = 25 ^oC.

Using the above data in the model of Figure D.9, four air penetration simulations cases were tried, using the following additional data for each case:

Simulation Cases	**Diameter Ratio D_1/D_2[a]**	**Supplied Pressure at the Inner Tip of the Immersed Pipe[b] KPa**
Case 1	0	101.7
Case 2	11.22	107
Case 3	17.72	115.0
Case 4	25.89	130

[a] D_1= funnel diameter, D_2= pipe diameter, [b] Calculated using Bernoulli equation D.23.

Table D.1: Data supplied for each simulated cases.

The results of these cases are shown in Figures 4.26, 4.27, 4.28 and 4.29, for cases, 1, 2, 3, and 4, respectively, showing air distribution inside the simulated porous medium of the biofilter. However, these simulation case should be taken as an indicative results of air distribution inside a porous medium, using an injection pipe system, in which results depended greatly on the characteristics of the soil.

D.8 Summary

The key factors for good performance of landfill soils have been identified and well researched. These key factors, particularly those of the soil characteristics such as porosity, permeability, thermal conductivity, hydraulic conductivity and other groups of factors, are important for the enhancement of landfill gas oxidation. Moreover, the importance behind them stem from the fact that they can be controlled to suit individual landfill circumstances while staying within the regulations limits. Nevertheless, there is still much work to be done to transform this well-found knowledge and information into actual technical design concepts. Attempts to propose technical systems to mitigate methane and other gases emanating from landfills have produced minor successes, in which none of these proposed concepts has ever been adopted in consideration of prescribed regulations up until now. However, a major drawback of all of these proposed passive systems is that they all have failed to have a mechanism to deliver oxygen to the soil's microcosms in a sufficient and sustainable way. Each of these systems is dependent upon the natural diffusion of oxygen into the soil via the molar, atmospheric, and wind surface pressures, which could only penetrate into few centimetres of the surface of the landfill cover, leaving the rest of the layers without oxygen. Natural oxygen diffusion is unpredictable, inefficient and is neutralized or overpowered by the upflowing pressure of the landfill gases. Since oxygen is an essential and a detrimental element in the mitigation of methane, as well as of the other gases in landfill soils, it will not contribute to reaching an optimum landfill cover if it is not supplied in a controlled, sufficient, sustained, and in a predictable manner.

In an attempt to alleviate these drawbacks, a new concept design has been proposed that would use the wind power to drive air (oxygen and nitrogen) into the layers of the soil, in a passive way, requiring no energy input, and would need no or little maintenance. The concept is simple in design, uses low technology, and easy to construct and assemble. The system is flexible and has many variables that could be adjusted to suit each particular demands and conditions of the site. In terms of the actual design, the system has a gate valve that allows air to flow in and prevent the landfill gases from escaping. It also has a flexible concentrating air-gathering cone to allow wind energy to produce higher pressure,

which is able to overcome the pressure of the up-flowing gases in the landfill, and deliver the air to the lower layers. In addition, the sizes of the system parts are freely controllable so that they could fit the needs of each individual landfill site. The key quantities of the system, such as the pressure gradient and the volume air quantity have been calculated, indicating that they depend only on the cross-sectional areas of the system and the prevailing wind speed at the site. These quantities are easily obtainable.

More importantly, this system is a new concept and offers a new research venue that needs substantial investment in time and effort, and eventually expected to obtain improved and higher performance levels compared to the other design concepts introduced so far, without violating any of the prevailing regulations.

Appendix E
Characteristics and Properties of cover Layer

Characteristics and properties of cover layer

E.1 Waste composition

Wastes discarded into landfills, their volumes, and characteristics vary greatly from one community to another and throughout the seasons of the year. Wastes could contain more construction and solid materials in one site; while they could have more biological contents in another site, and could have varying composition of materials in between other sites. Diversity of waste depends on the geographical locations of the landfills among the wide social communities across the globe. This variation in contents and in latitudes could have an effect on the temperature, moisture contents, and several other factors influencing the microenvironment of the landfill, that could sequentially affect the type of the degrading bacteria in that environment, consequently, resulting in the difficulty of characterising these bacteria and their behaviour (Asfari and Mashan'n, 2002).

Taking the United States (US), a highly developed country as an example, despite efforts at conserving, recovering, and recycling wastes, there still are reports on the ever-increasing amount of wastes discarded in its landfills and the generated municipal waste contents from 1960 to 2012, as shown in Table E.1 (US EPA, 2014). The US has produced an enormous 250.89 million tons of wastes, of which only 86.62 million were recovered and 164.27 million left buried in landfills in 2012. Those reported wastes constituted an increase from 88 million tons generated in 1960 to 250.89 million tons produced in 2012, indicating an average of 4.38 lbs (2.02 kg) of waste per capita a day. Out of those wastes, an approximate 70 million tons contained 57.7% yard trimmings and 4.8% foodstuff. From these waste contents, only 21 million tons were recovered for composting; while a mix of as much as approximately 58 million tons of organic materials (foodstuff, tree discards, and other organic materials) were left to the landfills, showing a 34.5% high rate of recovery from all generated wastes in 2012. The table also shows a high rate of recovery, reaching 33.3–34.7% in the last four years of the statistics, indicating a constant rate of recovery for both materials

and other wastes, suggesting that the maximum rate of recovery had been achieved (USA EPA, 2014).

Materials	1960 (%)	1970 (%)	1980 (%)	1990 (%)	2000 (%)	2005 (%)	2008 (%)	2010 (%)	2011 (%)	2012 (%)
Paper and Paperboard	16.9	15.3	21.3	27.8	42.8	49.5	55.5	62.5	65.6	64.6
Glass	1.5	1.3	5.0	20.1	22.6	20.7	23.1	27.1	27.6	27.7
Metals										
Ferrous	0.5	1.2	2.9	17.6	33.1	33.0	33.4	34.3	33.0	33.0
Aluminum	Neg.	1.3	17.9	35.9	27.0	20.7	21.1	19.4	20.5	19.8
Other Nonferrous	Neg.	47.8	46.6	66.4	66.3	68.8	69.1	69.3	68.5	68.0
Total Metals	0.5	3.5-	7.9	24.0	34.8	34.3	34.7	35.1	34.2	34.0
Plastics	Neg.	Neg.	0.3	2.2	5.8	6.1	7.1	8.0	8.4	8.8
Rubber and Leather	17.9	8.4	3.1	6.4	12.3	14.4	16.8	17.7	17.8	17.9
Textiles	2.8	2.9	6.3	11.4	13.9	15.9	15.4	15.3	15.4	15.7
Wood	Neg.	Neg.	Neg.	1.1	10.1	12.4	13.7	14.5	14.9	15.2
Other **	Neg.	39.0	19.8	21.3	24.5	28.2	27.5	28.3	28.3	28.3
Total Materials in Products	10.3	9.6	13.3	19.8	29.7	32.0	34.1	36.6	37.6	37.0
Other Wastes										
Food, Other (electrolytes in batteries and fluff pulp, feces, etc.)										
Yard Trimmings	Neg.	Neg.	Neg.	Neg.	2.2	2.1 6	2.3 6	2.7 5	3.5	4.8
Miscellaneous	Neg.	Neg.	Neg.	12.0	51.7	1.9	4.7	7.5	57.3	57.7
Inorganic	Neg.	Neg.	Neg.	Neg.	Neg.	Neg.	Neg.	Neg.	Neg.	Neg.
Total Other Wastes	Neg.	Neg.	Neg.	6.8	25.4	29.9	31.1	27.6	27.8	28.7
Total MSW Recovered -%	6.4	6.6	9.6	16.0	28.5	31.4	33.3	34.0	34.7	34.5

Table E.1: Percentage of municipal waste contents generated in the United States of America in 2012 (Source: US EPA, 2014)

In contrast, some developing nations have different municipal waste contents and mixes. Table E.2 shows typical waste composition of some of these developing countries (Asfari and Mashan'n, 2002), indicating a high rate of organic matter in waste generated relative to metals, glass, plastics, and paper. The lowest organic makeup of the waste was reported to have been generated from the United Arab Emirates (UAE), showing a rate of 42% waste with the highest at 63% from Jordan and Iraq; however, the highest per capita waste generators are the UAE at 770 kg/yr, and Kuwait at 660 kg/yr. Conversely, USA for recovery practices, as shown previously, the recovery rates in these countries are very limited; wastes from yard trimmings and trees are almost non-existent, due to the climatic nature of limited water in these countries.

Countries	kg/capita/yr	Organic Matter (%)	Paper (%)	Plastic (%)	Glass (%)	Metals (%)
Bahrain	584	59.1	12.8	7.4	3.4	2.1
Iraq	285	63	1	1	1.6	1.1
Jordan	330	63	11	16.8	2.1	2.1
Kuwait	660	51	19	13	4.5	5
Lebanon	220	59	18	8	8	2.4
Oman	256	60	8	12	10	9
Qatar	475	57	18	12	3	5
Syria	185	62	4	7	4	6
(Dubai) Abu Dhabi	750 542	42 49	6 6	10 12	3 9	3 6
Yemen	165	55	14	13	1.5	2

Table E.2: Percentage waste composition in some developing countries (Source: Asfari and Mashan'n, 2002)

Solid wastes' makeup could affect the holding capacity of water in landfills, and, in turn, the degradation process of the wastes. The increase of media aggregates, such as tree branches, construction materials, glasses, and plastics, etc., could affect methane production, depending on the types and sizes of these aggregates. Conversely, when the materials of the solid wastes, such as compost, wood fibres, and peats with high structural properties, are amended in the top cover soil of the landfill, they can help out in maintaining continuous oxidation, and prevent pore clogging, (Streese and Stegmann, (2003, 2005). Then again, these amended high aggregate sizes could increase the holding capacity of water filling in the voids and overwhelming the bacteria, thus, depriving them of oxygen. In addition, the type of waste media, particularly the quantity and availability of organic materials, if not stable, could aerobically degrade through chemical reactions. Such reactions could result in the oxygen, internally stored in the voids and spaces created by initial waste filling, being consumed, as well as producing other organic by-products in the process, subsequently,

competing with the bacteria present in the landfill. Moreover, demolition and construction materials buried among landfill wastes could also have an effect on the quality of the waste and the type of gases produced from landfills. Construction materials, such as gypsum boards that contain sulphides, and when present among organic material and in the presence of moisture, could produce hydrogen sulphide gas in an amount that could reach from 50 to 15000 ppm (USA EPA, 2010). This gas can have a serious and harmful effect on the environment. These are merely some examples, indicating the effect of waste and its composition on the activities of bacterial communities when deposited in landfills, although varying greatly from one site to another all over the globe. This variation of waste materials in landfills, pairing along with social communities, poses a difficult challenge to environmentalists and concerned governments alike.

To contain these wastes from contaminating the surrounding environments, covers are placed on top of these discarded wastes to prevent human contacts, to allow vegetation growth, and to control the amount of gases emitted from the landfills. Many prevailing landfill regulations however, require that the top landfill covers be mostly of low permeable materials to allow limited amount of rainwater to migrate into the wastes. In essence, landfill sites are essentially reactors in the presence of oxygen and by the action of the microorganisms when the moisture is mixed with organic materials, methane gas, carbon dioxide, and leachate runoffs are produced. This continuous reaction in the landfill layers occurs specifically in the top landfill covers, where the characteristics of these covers provide the favourable conditions for these methanotrophic bacteria to flourish.

E.2 Cover layer thickness

The top layer of the landfill is where most of the methane assimilation takes place. An increase in layer thickness can lead to augment the rate of methane oxidation and can also increase the capacity of the layer to hold both methane and oxygen much longer together. On the other hand, an increase in the thickness of the layer would increase the cost of landfill closure. Additionally, a preferred cover layer design, usually with specific material type to encourage further oxidation would require additional costs in transportation, construction

materials, distribution, labour, etc., adding up to the overall cost of closure. Moreover, increase in cover thickness could also aggravate leachate pollution, due to the increase in water holding capacity for such capping system (Huber-Humer and Lechner, 2003). The recommended degrees of thickness suggested by Huber-Humer (2004) and Martikkala and Kettunen (2003) were 120 cm and 40–50 cm, respectively. Other important cover elements, such as soil grain sizes, porosity, and water holding capacity of the layer supplement the thickness characteristic.

E.3 Cover density and compaction

Methanotrophs present in the soil require sufficient oxygen to oxidise methane. Pores and gaps present in the cover soil are the sources of this oxygen, and when a soil layer is compacted by action of heavy machineries or natural settlements, oxygen is squeezed out of these pores. Therefore, it is recommended that the density of the cover layer should not exceed 0.8–1.1 T/m^3 in order to have an optimum oxidation level (Huber-Humer et al., 2009). Unfortunately, however, this density level or any other specific density throughout the cover layer is practically unattainable due to continuous settlements, rainwater saturation, and the creation of continuous voids due to decompositions.

E.4 Soil grain size

Methanotrophic bacteria attach themselves via a polymeric substance to the grains of the soils in the cover layer of the landfills (Hanson and Hanson, 1996). In theory, finer grains allow more surface areas for methanotrophs to multiply and exist in larger numbers, therefore, providing greater chances for these bacteria to oxidise methane. Correspondingly, according to Streese and Stegmann (2003), higher oxidation rates are achievable in finer grain sizes in compost cover material than in the coarse grains medium. Conversely, oxidation rate of methane decreases rapidly to the minimum in the finer medium. This behaviour could be due to the rapid clogging of the fine pores in between the grains of the medium and to the high retention characteristic of finer grains of water. Other studies have reported that oxidation is higher and steadier in coarse grain sizes cover layers than in the

finer grains (Boecks et al., 1997), which may be attributed to the sizes of the pores, obtaining more oxygen and moisture, hence, sustaining oxidation. Bender and Conrad (1995) suggested a range of 50 µm to 2 mm in diameter of soil medium to be the optimum aggregate size makeup of the cover layer. In more recent studies, Huber-Humer (2004), Huber-Humer et al. (2008, 2009), and Scheutz et al. (2009b) suggested that the best cover layer in order to achieve a balance between water retention, oxygen, and gases filtration ratios toward providing a stable organic and methane oxidation reaction is a cover layer that is made up of mature compost with coarse grain sizes. Barlaz (2004) on the other hand, argued that compost could produce methane under anaerobic conditions if less oxygen is available in the soil. It can also inhibit methanotrophic bacteria in the presence of high nitrogen and ammonia, when both interact together with the limited available oxygen, therefore, competing with CH_4 oxidation (Bodelier and Laanbroek, 2004), i.e., if the compost has not been matured enough. Thus, caution is called for when using compost material with any type of grain sizes for landfill covers.

E.5 Porosity

Porosity is defined as the ratio between the volumes of the voids in these reactive layers to their total volume (Klausner, 1991). It is another important element to consider when referring to characteristics of the cover layer. A distribution layer normally placed just underneath the cover layer is where oxygen and methane interact with each other and with the other elements in the cover layer. Pore sizes in both of these two layers allow water and gases to exchange freely within the media, consequently, delivering sufficient supply of all of the elements to the methanotrophs. Through the property of porosity, methane is transported from waste layer below to react to the distribution and oxidation layers above, in an advective flux action driven up by the gas production pressure (Kjeldsen, 1996). Oxygen is transported to the oxidation layer by diffusion from the atmosphere.

Porosity of the cover soil changes and decreases with time, due to the degradation of the material in the oxidation layer, that is, if the cover material is made up of organic materials or amended with organic materials. An example is the yard waste compost, where the density

increases from 600 g/L to 1000 g/L after only 218 d in a column experiment. This, in turn, decreases the pore sizes in the material followed by a decrease in the porosity of the cover layer in a considerable amount (Philopoulous et al., 2009). Scheutz, et al., 2009a in another research reported that cover soil has the potential to compact and settle to a size of more than 20% of its original state. To remedy this action, according to Philopoulous et al. (2009), would be to have an amended cover soil with other materials, such as sand and perlite that would stabilise the layers and increase the grain sizes of cover materials, eventually, generating an interdependent relationship between grain sizes and porosity.

Porosity, being a complex concept, has been given several definitions. Total porosity is defined as the total volume of voids over total volume of the layers. This is a concept that is misleading and could lead to skewed calculations, considering that not all voids of layers could contribute to the process of oxidation, if they are isolated or unconnected. Alternatively, other porosity definitions and calculations have been proposed. One of these definitions was proposed by Olivier and Gourc (2007) and referred to as the open porosity, given as—

$$n_o = 1 - V_s/V \qquad \text{E.1}$$

where,

n_o= open porosity,

V_s= total volume of the solid in the layers,

V= total volume of the layers.

From this equation, Olivier and Gourc calculated open porosity of municipal solid waste and found it to fall between 48 and 51% of landfill waste, which are of high levels. Another porosity concept has been put forward by Hudson et al. (2004), proposing another term called effective porosity and is given by the equation as follows:

$$n_e = n - (V_g + V_w)/V \qquad \text{E.2}$$

where, n_e= effective porosity,

n= total porosity,

V_g= volume of gas remaining in spaces of the pores of the layers,

V_w= Volume of liquid (water) remaining in pore spaces and cannot be drained.

They calculated that the effective porosity of municipal solid waste according to this concept is to fall between 1.4 and 14.4%, in comparison to the total porosity, which was found to range between 45.5 and 55.5%.

The difficulty of calculating porosity springs from the time settlement of the landfill itself, alongside with calculating the voids and solid volumes in landfill layers, even when taking averages of grain sizes and volumes of voids from core samples of landfill sites. When the weights of the layers press down on each other, the compressible objects in the landfill layers collapse on themselves, filling in the void spaces in the layers in a fashion described by Grisolia et al. (1995) in Figure E-1. This would infer that since porosity is a dynamic concept, calculating its values could change with time, effectively affecting oxidation in a dynamic and variable way.

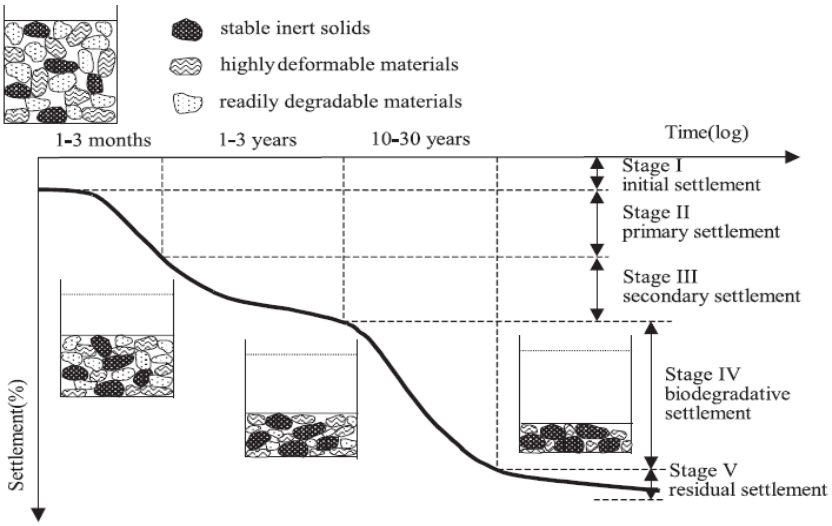


Figure E.1: Settlement due to layer pressures (source: Grisolia et al., 1995).

E.6 Water retention and holding capacity

Microbial activity in the soil is affected by another essential characteristic of the distribution and reaction (oxidation) layers. This characteristic is the ability of the layers to hold and keep

sufficient amount of water over time to sustain a continuous interaction between methane and oxygen. This property is a balance between the media's water holding capacity and its porosity. Conditions, if made right would allow sufficient movement of oxygen and methane for maximum methane consumption. Therefore, media's water holding capacity could be a measure of the granulometric composition of the distribution and reaction layers to retain moisture.

Water holding capacity is another important element from among the set of characteristics of the cover medium and is difficult to calculate, as well. As of yet, little research to provide understanding and quantification on the subject has been done, except for Huber-Humer et al. (2009) who recommended favourable values of water holding capacity for a compost material as a cover layer, proposing a range of 50–130% of dry matter (DM) for the capacity values.

E.7 Permeability and hydraulic conductivity

Hydraulic conductivity is the measure of the degree of the penetration of external water, such as rain or water runoffs into the cover layers and its infiltration into the distribution and reaction layers beneath. In this process, water drains into the soil medium, and consequently affects the moisture content of the medium, and which in turn affects the movement of liquids and gases within the medium of the layers. This action therefore, provides or prevents essential nourishments to the bacteria within the layers. In essence, hydraulic conductivity is a measure of the cover layers' granular makeup, which depends on grain sizes, grain orientations, soils' porosities, permeating liquids viscosities, degree of saturation, density, and porosity of the medium. Generally, hydraulic conductivity is measured vertically; although, it could have horizontal values as well, mostly with different values, as often, soils are not homogenous in nature.

Measuring hydraulic conductivity is a complex process. Cassiani (1998) divided the work of calculating hydraulic conductivity into three categories. The first is based on Zangar's (1953) measurement, which involved analytical solution with approximate boundary conditions. In

this calculation, capillary effects were neglected. The second is a measure based on Philip's (1985) model, which is another analytical calculation that considered capillary effects. The third is the numerical model calculation that can include boundary conditions in a more accurate representation, and can have the flexibility to take capillary effects into consideration (Stephen and Neuman, 1982). Each of these measuring models has some advantages and some drawbacks. Differences and advantages/disadvantages of all of these models are discussed by Pradeep et al. (2006). While there were other models, they were mostly intended for hydrological soil investigation tests. One of these other models is the Kozeny-Carman model (Mitchell and Soga, 2005), which is complex and based on empirical representation that requires the calculation of a multitude of experimental factors. The model takes into account earth's gravity, viscosity, pores' shapes, wetted surface area of the particles, void ratio, and the degree of saturation. One of the most widely accepted formulations for calculating conductivities however, is Darcy's law (1856), and widely used because of its simplicity. It states that the total liquid discharge through porous medium is proportional to the pressure drop through that medium. Although Darcy was the first to state this relationship, which was founded on experimental trials, the law was based on the conservation of momentum and could be driven from the basics of fluid mechanics, such as the Navier-Stokes equations.

Reddy et al. (2009) reported hydraulic conductivities from the literature for field tests, in which hydraulic conductivities have been suggested as a range of values rather than as exact and finite values (Table E.3). These suggested ranges were proposed in that manner because of the difficulty in measuring and quantifying conductivities in soils. Unit weight (specific weight) refers to the weight of soil per unit volume of the same soil calculated as dry or wet.

Hydraulic conductivity is not constant for soils. As time evolves, interconnected soil voids in relation to grain sizes change with the settlement of the soil and with the amount of precipitating materials within these voids. In addition, the ions present in the permeating liquid interact with the minerals present in and on the grain surface's particles; hence, the accumulated salts fill in the spaces between these voids, thereby affecting the rate of flow through the voids (Aringhieri and Giachetti, 2001). One of the most common materials in the

soil is carbonate salt (VanGulck et al., 2003). Another factor that affects hydraulic conductivity with the passing of time is the bio-clogging factor (Vandevivere, 1995), which results from the accumulation of materials due to bacterial activities in and around grain voids in the soil. Francisca and Glatstein (2010) determined the long-term hydraulic conductivity of compacted soils haing biological activities. The soil that they used consisted of compacted silt and silt-bentonite mixture. On the other hand, bio clogging due to bacterial and yeast activities had significant effects on the lowering of soil conductivity by a factor of 10^2, using either distilled water or nutrients solution (Francisca and Glatstein, 2010; Vandevive, 1995; Clement et al., 1996; Seki and Mlyazaid, 2001; Thullner et al., 2002). This has been attributed to the formation of a bio-film inside the reaction layers. Figure E.2 shows an interesting relationship between different empirical models and an experimental result, which showed good agreement.

Reference	Source	Unit Weight (KN/m^3)	Hydraulic Conductivity (cm/s)
Landva and Chark, 1986	*In situ* test pits, Calgary *In situ* test pits, Edmonton *In situ* test pits, Mississauga *In situ* test pits, Waterloo	12.5–14.5 10.0–12.9 10.7–13.6 10.5–13.1	$2.6x10^{-2}$–$1.6 x 10^{-2}$ $1.3x10^{-2}$–$1.1 x 10^{-2}$ $5.0x10^{-3}$–$1.0x 10^{-3}$ $1.3x10^{2}$–$1.1 x 10^{-2}$
Ettala, 1987	Modified double cylinder infiltrometer and pumping tests	Heavy compaction Slight compaction	$2.5x10^{-6}$–$5.9 x 10^{-7}$ $2.5x10^{-5}$–$2.0x 10^{-5}$
Oweis et al., 1990	*In situ* pump test *In situ* falling head test* Test pit infiltration	–	$1.0x10^{-3}$ $1.6x10^{-4}$ $1.3x10^{-3}$
Shank, 1993	Slug test, 20- yr- old MSW	–	$9.8x10^{-4}$–$6.7 x 10^{-5}$
Jain et al., 2006	Borehole permeameter test	3- to 6-m depth 6- to 12-m depth 12- to 18-m depth	$6.1x10^{-5}$–$5.4 x 10^{-6}$ $2.3x10^{-5}$–$5.6 x 10^{-6}$ $1.9x10^{-5}$–$7.4 x 10^{-6}$

*Falling head is a technique allowing water to fall under gravity pressure.

Table E.3: Field hydraulic conductivities for municipal solid waste, with dry unit weights, as a characteristic of the waste (Source: Reddy et al., 2009)

Another equally important factor, beside bio clogging, is the pressure effects of layers on hydraulic conductivity. Powrie et al. (2000) indicated that a linear rapid relationship between vertical pressure and soil hydraulic conductivity does exist. Figure E.3 shows this

relationship, in which conductivity decreases, as the pressure of the above settling layers increases.

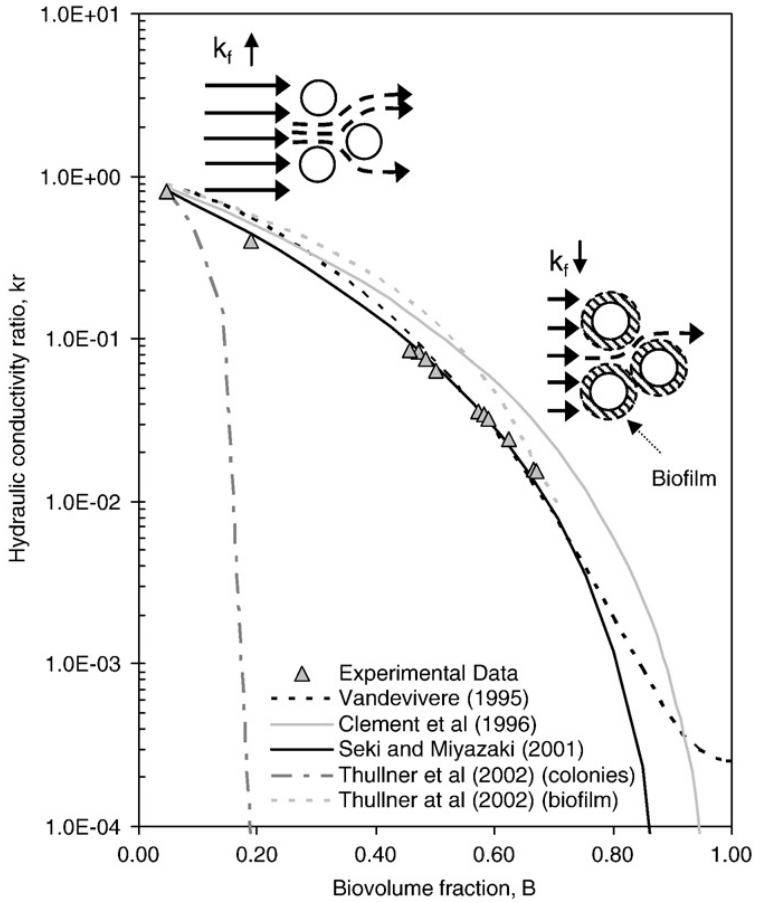


Figure E.2: Different models describing the effects of biological activities on hydraulic conductivity (source: Francisca and Glatstein, 2010). (Conductivity ratio is the ratio of the final conductivity after bio buildups and the initial soil conductivity; while bio-volume fraction B is the ratio of void space change due to bacterial activity, with respect to the initial void space).

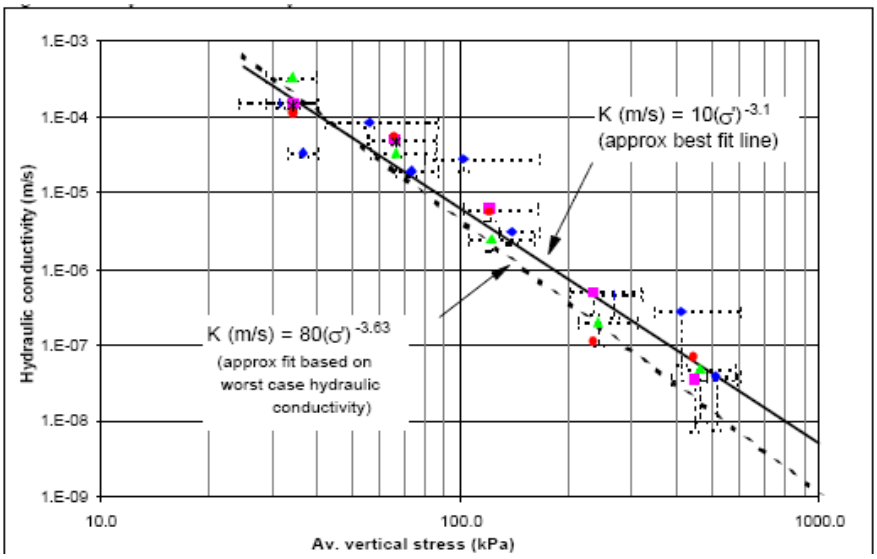


Figure E.3: Effects of vertical pressure on hydraulic conductivity (source: Powrie et al., 2000).

While hydraulic conductivity is the measure of the degree of penetration of external water into the soil, permeability is the soil's resistance to fluid flow (air or water) and is the measure of pore sizes, grain sizes, grain distribution, and type of soil structure. It is primarily a function of void ratio, and is regarded as one of the most important soil characteristics among all of the other sets of soil characteristics, affecting the various activities of methanotrophic bacteria. The soil, in allowing fluids to pass through its structure, has a direct bearing on air diffusion, water percolation into the layers of the soil, and leachate seepage. Permeability in terms of mathematical science is the proportionality constant between fluid speed, viscosity, and the pressure gradient through the soil, and described by relationship according to Darcy's law, as follows:

$$v=(k/\mu)(\Delta P/\Delta x), \qquad \text{E.3}$$

where,

v= the superficial velocity (the velocity of the fluid, in the assumption that the fluid is the only phase present in the medium) in m/s.

μ= the dynamic viscosity of the fluid, in Pa s.

k= the intrinsic permeability (or the absolute permeability) is the hydrodynamic permeability of the porous soil and is independent of the type of fluid permeating through, which is dissimilar with the hydraulic conductivity; the hydraulic conductivity is a measure of both liquid and soil properties, measured in m^2 (or in Darcy's units).

ΔP= pressure applied across the medium section, in Pa.

Δx= the distance of the path travelled by the fluid, in m.

Equation E.3 can be expressed in terms of the permeability as follows:

$$k = v\,\mu\,(\Delta P/\Delta x) \qquad \text{E.4}$$

As aforesaid, intrinsic permeability is the measure of the property of the soil itself, and has a different meaning from hydraulic conductivity; however, both coefficients are related through the equation as follows:

$$k = K\,\mu/\rho\,g, \qquad \text{E.5}$$

where,

k= intrinsic permeability, m^2 (Darcy),

K= hydraulic conductivity, in m/s,

ρ= the density of the fluid, in Kg/m^3,

g= the gravitational acceleration of earth at the site, in m/s^2.

The intrinsic permeability, as a measure of the property of soil, is constant for normal soil structure, but it could change in case of landfill soil settlements. Accordingly, when calculating permeability, this fact of settlement has to be taken into account.

In real landfill fluids interaction, both water and air are present at the same time in each of their varying degrees of saturation. Therefore, two different permeabilities exist simultaneously in the soil structure, namely, gas permeability, k_G (effective permeability of

gas), and water permeability, k_L (effective permeability of liquid), with the presence of one phase inhibiting the other, depending on their relative permeabilities. Accordingly, the relative permeability of both phases could be expressed in terms of intrinsic permeability as in the following:

$k_{rG}=k_G/k$, E.6

$k_{rL}= k_L/k$, E.7

Both of these values are dimensionless coefficients and always <1.

For the saturation of each phase alone, the relative permeability becomes equal to 1; hence, the effective permeability will be equal to the value of the intrinsic permeability of the soil. Figure E.3 describes the interaction between the two permeabilities of a two-phase flow of water and gas in a soil structure, in which one permeability runs counter the other. Warrick (2001) stated that also at saturation, the relative permeabilities (S_{rG}, S_{rL}) reach 1 at the end of the phase spectrum.

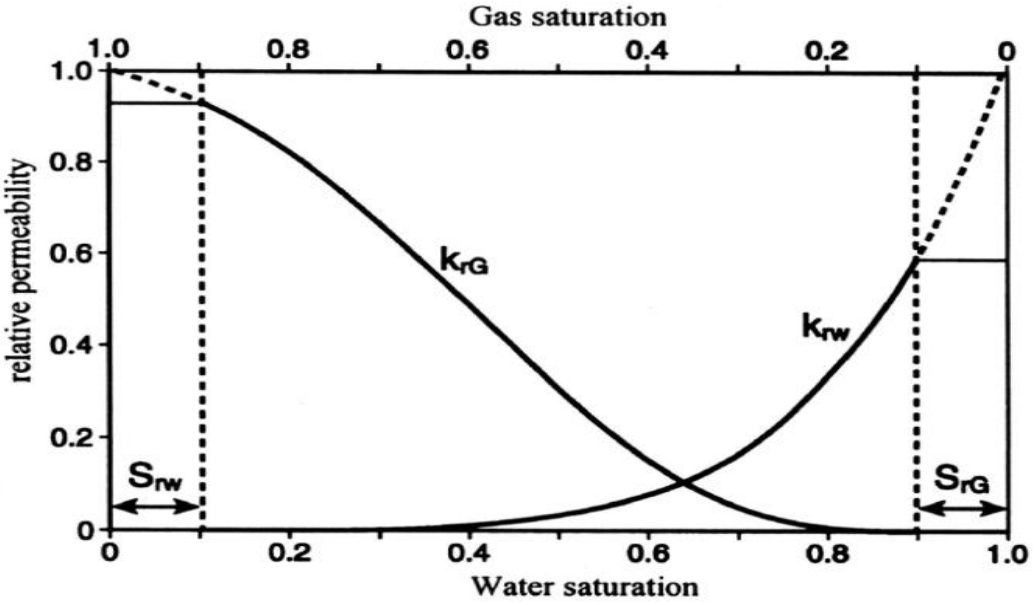


Figure E.4: Two- phase relative permeability of gas and water in the soil structure (source: Warrich, 2001).

Darcy's formula, equation E.3, is intended for use in a steady state flow of a homogenous medium, which is not the case for waste-compacted soils, where the porous waste medium is composed of different materials with diverse nature. Darcy's equation can be used in that case as an approximation calculation model. Other researchers have suggested the use of other approximation to determine soil permeability by empirical approach (Hazen, 1911; Klinkenberg, 1941; Massmann, 1989), experimentally (Scheidegger, 1974; Corey, 1986; Sharp et al., 1994; Springer et al., 1995), and by the use of numerical methods (McDonald and Harbaugh, 1988; Celia and Binning, 1992; Joss and Baehr, 1995).

E.8 Heat capacity and thermal conductivity

Thermal conductivity is a characteristic of the composition of the soil within the layers. This characteristic describes the ability of the soil to absorb or emit heat through its surface and is a temperature-dependent property. From a mathematical viewpoint, thermal conductivity is a constant of the proportionality between the amounts of heat flowing through materials and the temperature gradient (Callister, 2003). A general formulation of the generated heat in one direction, for a uniformly isotropic media, is given in an equation as follows:

$$Q = - k A (\partial T/\partial x), \qquad \text{E.8}$$

Where, Q= total heat generated in units of heat watts,

k= thermal conductivity in units of heat per unit length per temperature units in Kelvin ($W/m.^{o}K$),

A= bulk cross-sectional area, unit area (m^2),

$\partial T/\partial x$= temperature gradient is in units of temperature per unit of length ($^{o}K/m$).

The formulation is a direct application of the second law of thermodynamics, and is a formulation analogous to that of Darcy's law, stated in the aforementioned Equation E.7. Thermal conductivity for isotopic media is uniform throughout the media and in all directions; however, for heterogeneous materials, such as the soils of landfills, thermal

conductivity can take several values in all of the directions and at each of the different levels in the soil, making calculations of this heterogeneous material a difficult undertaking. The difficulty in quantifying thermal conductivity for waste materials in landfills stems from the large number of variables to contend with, all at the same time. Solid particles, liquid, and air have each its own thermal conductivities with varying densities and varying degree of contacts at varying degrees of water saturations (Ahn, et al., 2009; Chandrakabthi et al., 2005).

Microorganisms in landfills produce heat due to their activities in consuming the organic materials present in the waste. The heat will accumulate and increase substantially if the heat is not dissipated uniformly. Methanotrophic bacteria need a balanced environment in which the temperature must remain in the range of 25–35°C (Whalen et al., 1990). The landfill cover must have the characteristic to breathe efficiently. On the microscale level, heat is dissipated by grain-to-grain contacts, or through grain-to-water contacts, all by conduction process, and grain-to-air, trapped in voids by convection process, depending on the layer's micro arrangements. Thereafter, all the heat could dissipate through the upper layer by way of these processes in addition to the radiation process. Because thermal conductivity of the air is much less than that of liquids or solids, an aerated soil of having more air-filled voids will produce lower thermal conductivities than that of the soil with filled voids of solid particles or liquid. It is best that a desirable cover material must have the characteristic of holding and emitting enough heat throughout the layers to maintain a favourable temperature range suitable for methanotrophic environment. In the literature, biologically stable compost cover material is favoured for its quality of having better methane oxidation rate due to its balanced air/moisture and breathing characteristic (Huber-Humer, 2004), despite its other drawbacks. Compost material's conductivity depends directly on its density, grain/void ratio, water holding capacity, and its heat capacity. Heat capacity is the ability of a material to acquire a measurable amount of heat when temperature is changed by a given amount in joule per temperature degree ($J/^{o}K$). Although little was done to quantify thermal conductivity of landfill wastes, Akn et al. (2009) was able to study the thermal conductivity, heat capacity, and thermal diffusivity of 12 compost bulking materials (Table E.4). They concluded that for an 80% of water holding capacity,

compost material, sawdust, soil compost blend, beef manure, and turkey litter have shown the highest thermal conductivities and heat capacities, at ranges of 0.12–0.81 W/m°C and 0.93–3.09 Mj/m^2°C, respectively.

Experiments on compost materials, as a top cover, have been reported in the literature to have met considerable success (Huber-Humer, 2004; Scheutz and Kjeldsen, 2003). Nonetheless, little has been done to quantify the conductivities of landfill soils. However, because it is imperative to provide a suitable thermal environment for methanotrophs to function properly, and also given the difficulty to calculate waste and soil conductivities, thermal conductivities of the surrounding grounds are sometimes taken to be an indicator of the thermal conductivity of landfill soil. Table E.4 shows some thermal conductivities of different materials and soils. Unfortunately, however, thermal conductivities of cover materials used on landfills in Kuwait are not available.

	Thermal conductivity (W/m °C)			Volumetric heat capacity (MJ/m^3 °C)			Thermal diffusivity (mm^2/s °C)		
	Dry	80% WHC	S[a]	Dry	80% WHC	S	Dry	80% WHC	S
Wheat straw	0.02–0.07	0.03–0.17	0.53	0.03–0.99	0.26–1.07	4.13	0.07–0.57	0.11–0.29	0.13
Sawdust	0.03–0.05	0.17–0.47	0.44	0.29–0.49	1.39–1.70	3.91	0.10–0.11	0.12–0.27	0.11
Soil compost blend	0.06–0.12	0.16–0.81	0.59	0.84–1.44	1.41–1.89	2.94	0.07–0.12	0.11–0.50	0.20
Silage	0.03–0.09	0.09–0.47	0.53	0.17–1.75	0.93–1.76	3.92	0.05–0.16	0.09–0.27	0.13
Beef manure	0.03–0.08	0.17–0.52	0.39	0.21–0.96	1.66–2.50	3.82	0.08–0.14	0.10–0.21	0.10
Oat straw	0.02–0.06	0.05–0.18	0.56	0.06–1.07	0.40–1.09	4.08	0.06–0.35	0.09–0.38	0.14
Soybean straw	0.02–0.07	0.06–0.30	0.54	0.05–0.99	0.28–1.32	4.10	0.07–0.44	0.09–0.30	0.13
Cornstalks	0.02–0.05	0.03–0.24	0.53	0.05–0.60	0.46–1.44	4.09	0.08–0.40	0.07–0.28	0.13
Alfalfa hay	0.03–0.05	0.07–0.15	0.43	0.05–0.82	0.37–1.54	4.09	0.07–0.55	0.09–0.33	0.11
Leaves	0.02–0.08	0.06–0.38	0.64	0.04–0.90	0.23–1.86	4.12	0.07–0.47	0.05–0.97	0.16
Wood shavings	0.03–0.06	0.07–0.15	0.55	0.17–0.85	0.77–0.99	3.92	0.07–0.18	0.09–0.16	0.14
Turkey litter	0.03–0.08	0.12–0.50	0.51	0.37–1.11	1.57–2.58	3.62	0.07–0.09	0.07–0.20	0.14

[a] at saturation

Table E.4: Thermal conductivities, heat capacities, and diffusivity of several compost additives (Source: Ahn et al., 2009)

E.9 Diffusivity

Oxygen is an essential element for methanotrophs to assimilate methane and produce energy. A suitable medium of delivery is therefore required in order to deliver oxygen to the bacteria at the lower layers of the landfill. It is thus important for the oxidation layer to allow enough and continuous supply of oxygen to reach the microorganisms. Diffusivity is a coefficient referred to in Soil Science as the proportionality constant between the diffusion flux and the gradient of the diffusing material and is dependent on gas density and the prevailing pressure in the medium. Diffusivity, from the perspective of Soil Mechanics Science, is the ability of gas to move into the soil and the soil's subsurface with time, with units of square length per time. The total amount of binary air gas mixture (mixture of two components of gas molecules), diffusing into the soil in one dimension is given, Fick's Law of diffusion (Crank, 1980) as follows:

$$q=-D(\partial C/\partial x), \qquad \text{E.9}$$

where, q= diffusion flux in amount of substance per unit area per time;

D= diffusion coefficient; or diffusivity, in units of square length per time;

$\partial C/\partial x$= the concentration gradient, C, in units of substance per cubic length, per x, the space coordinate normal to the section, in units of length.

The total amount of air substance diffusing into the soil can be expressed by the following:

$$Q= -DA(\partial C/\partial x), \qquad \text{E.10}$$

where, Q= total air quantity, diffusing into soil in units of air substance per time (mol/s, or g/s).

D=diffusivity, (m^2/s)

A= – cross-sectional area of soil, with air passing through, (m^2);

$\partial C/\partial x$= the concentration gradient of air in direction normal to cross-sectional area, (mol/m^3 m, or g/m^3m).

This formulation is again similar to the equations of the hydraulic and thermal conductivities, described in the aforementioned equations E.7 and E.8, respectively.

Fick's law can take another form, in terms of time and gas concentration gradient. When employing the principle of mass conservation, the law takes the form as follows (Crank, 1980):

$$\partial C/\partial t = D\partial^2 C/\partial x^2 \qquad \text{E.11}$$

where, t is the time measurement for the gas concentration to diffuse through a medium in one directional space. This law is referred to as Fick's second law. For more than one molar gas type to diffuse into soil, Fick's law can take the form for binary gas mixture (Scheutz, 2009a) as follows:

$$q_i = - D_{ij}(\partial C_i/\partial x), \qquad \text{E.12}$$

with q_i= flux of ith gas component into the soil,

D_{ij}= the binary diffusion coefficient of the ith component in free gas mixture with the jth component,

C_i= the concentration of the ith gas in the gas phase,

x= the direction of gas movement in the soil.

The solution of these differential equations requires knowledge of the boundary conditions and the diffusivity to be constant all along the diffusion movements and across the medium length, which unfortunately, is not obtainable for the soil covers. Unlike thermal and hydraulic conductivities, diffusion coefficient, D in equations E.9, E.10, and E.11, is for binary mixture and is intended for use in a homogeneous medium with steady state diffusion, with only two diffusing components (binary model). The advective flux described in equation E.12 is an important component in calculating gas mixing and diffusion in soil, since it has been

found that 99% of methane generated to reach the oxidation layer is by advective flux (Molins et al., 2008). Other models to include several components, based on the Maxwell-Stefan model (Maxwell, 1866; Stefan, 1871) to approximate the diffusion coefficient (Wilke, 1950; Stein et al., 2001; Jakobson, 2008), have been also used.

Parallel to the well-presented theoretical and empirical models in the literature, which have been tried successfully, are the experimentation processes. These experimental processes are more commonly used to measure the diffusion coefficients of soils. As landfill covers and waste materials are made up of unique substances due to their inherent particular characteristics, experimental process becomes a necessary means to calculate the diffusivity that represents this uniqueness (Scheutz, et al., 2009a). An investigation of Allaire et al. (2008) on the five different laboratory methods to determine the diffusion coefficient of loamy sand gave conclusive results that the macro pores present in the layer structures pose as an important characteristic of soils, and that all five methods studied gave similar results in the absence of these macro pores.

The systems of diffusion models, based on Fick's law, are well-established and are being used extensively in the field of Soil Science. Conversely, if the binary gas diffusion coefficients were to be different by a factor of 2, these models would then produce inaccurate results (Leffelaar, 1987). In effect, other models have been proposed and used by researchers to simulate diffusion in soils such as empirical (Marrecro and Mason, 1972) and numerical models (Lui et al., 2006). One theoretical model worth noting is the Knudsen diffusion model. In this system, when gases are confined to diffuse in soil, a wall phenomenon takes hold; the grain boundary acts as a wall to limit free diffusion, depending on the size of gas moles. Free mean path of moles is limited by the pore sizes in the soil, which could affect diffusion, and hence, methane oxidation (Scheutz et al., 2009a). Molins et al. (2008) and DeVisscher and VanCleemput (2003) posited however, that Knudsen diffusion is not essential and can be ignored under optimum oxidation of methane, and is only regarded of importance when pore sizes are below 1 μm, in which case, and at ideal oxidation, these pores will, at any rate, be filled with water (Scheutz et al., 2009a).

E.10 Oxygen supply

Oxygen supply to the methanotrophs' microenvironment is a critical factor on methane consumption by these specific bacteria, without which could cause produced methane to escape into the atmosphere freely unreduced. In such a microenvironment, a counter gradient balance between methane and oxygen exists in that, oxygen concentration is reduced from high to low, moving from top of the oxidation soil to the bottom; while methane concentration is changing in the opposite direction, moving from high to low going upward. This kind of distribution is important, because it affects the microbial methane consumption process, as well as the vertical distribution of methanotrophs in the soil. Figure E.5 shows a typical counter gradient behaviour in the soil distribution of a landfill, as suggested by Scheutz (2002). However, this kind of counter occurrence in the soil for the gases is meant for normal landfill covers with free air diffusion. In contrast, for arid climates where dust fallout could precipitate continuously for most of the year, depositing an average of 278 tons per square metre (Al-Dousari, et al., 2014), and affecting surface permeability (Krishna and Suresh, 2016), free diffusion of air into the soil is hindered. It was suggested that the addition of 12.5% dust to the soil could limit soil permeability, particularly with the presence of moisture (Devaragan and Sasikumar, 2015). This kind of arid condition could otherwise be compared with the condition of waterlogging in temperate climates, where oxygen depletion could be found to be significant below 5 cm in waterlogged soils (Hanslin et al., 2005). This depletion of oxygen could be attributed to the interruption of the connectivity, joining the pores and void spaces together, when water molecules occupy these spaces forcing air molecules out, and consequently, decreasing the diffusion coefficient for air to enter into the soil. Reducing the presence of oxygen in the soil, due to dust fallout or in the condition of waterlogging, could affect bacterial type, presence, and activity. As well, it could also affect both the water uptake of the surface of the plant and its physiology. Nutrients, oxygen, nitrogen, organic matter, and all other mineral elements needed for biological metabolisms are all affected (Sairam et al, 2008).

Oxygen supply and its transport into the oxidation layers emanate from two sources; the first is given off from the air volume trapped into the soil when landfill soil was first constructed and trapped inside the voids and were forming cracks during the construction phase and layout of the landfill layers. This oxygen source of trapped air in the layers is voluminous; nevertheless, it is unsustainable, since it cannot be replenished again. This trapped oxygen in the soil, when consumed by the microorganisms producing carbon dioxide, other gaseous products, and organic materials, as shown in Equations 2.1–2.2, along with unreduced methane, would likely form an upward pressure in the layers, hence, limiting atmospheric air flux from entering into the soil. In addition, the trapped oxygen in the soil layers can only move through the soil and from one point to another in a flexuous way, only if the pore size and interconnectivity of these pores are in favourable alignments. The second source of oxygen supply comes from the diffusion of air from the atmosphere, in which air containing oxygen diffuses into the soil directly. The driving mechanism for this source comes from the fluctuating atmospheric pressure, as a pumping mechanism, by force of alternating winds, or by the direct molar difference of air diffusing into the top soil. In this latter case, air penetration into the soil is limited by the prevailing mechanism (Scheutz et al., 2009a). Free molecule flux or Keudsen diffusion flux mechanism, as previously discussed, occurs at the surface of the soil, only when the mean free path of the molecule (average distance of a gas molecule traveling before colliding with another molecule) is much greater than the pore size. In contrast, Warrick (2001) claimed that this mechanism is not as significant as the diffusion of air and oxygen due to air pressure caused by the prevailing winds and atmospheric pressures.

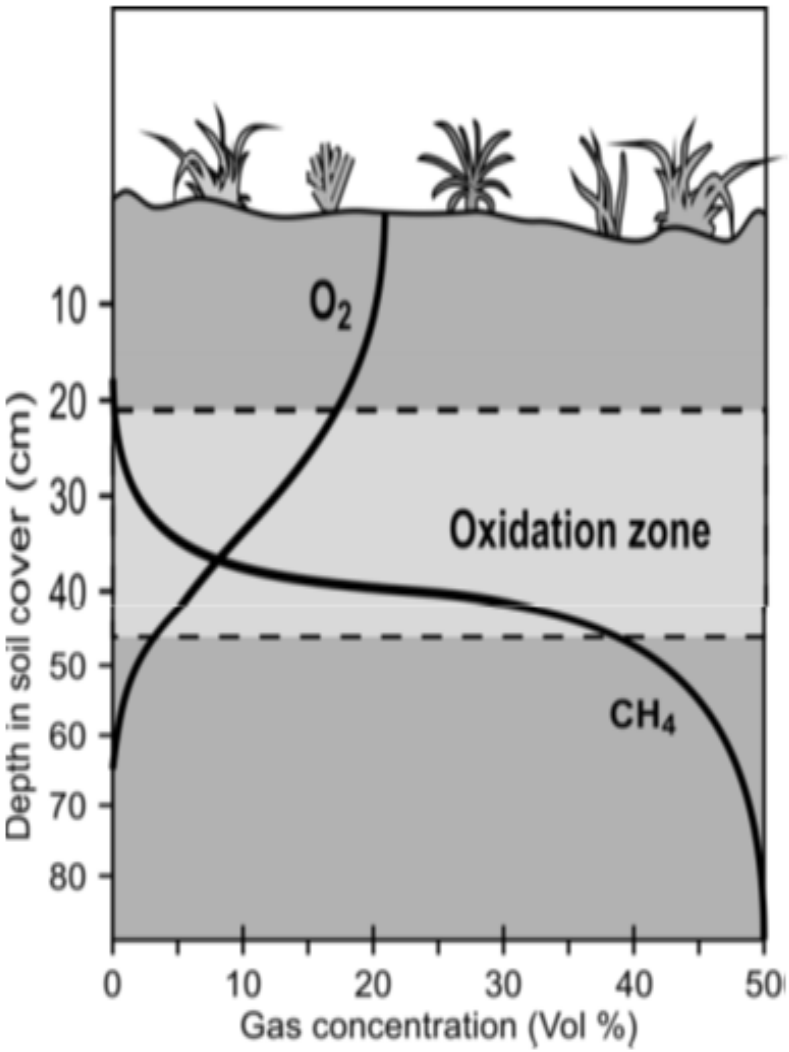


Figure E.5: Counter gradient of oxygen and methane in a typical landfill (source: Scheutz, 2002).

Meanwhile, Czepiel et al. (1996) found out that oxygen supply mechanisms combined can only supply oxygen to a maximum soil level of methane oxidation from 5 to 15 cm in depth of the cover layer. The same authors also reported that both low oxygen supply (intermittent supply) and low level of oxygen (oxygen volume supply) are responsible for the shallow oxygen penetration. This is somehow critical in that, the oxygen supply and its sustainability are vital in creating an interactive methane barrier in landfill soils. Huber-Humer (2004) confirmed through column tests that although lower oxygen supply can affect oxidation on

the short-term; albeit, methane oxidation can be consumed efficiently by the methanotrophic bacteria, only if oxygen quantity can be supplied continuously and sufficiently. While the maximum methane oxidation occurs at a depth of 5–15 cm, the range of oxidation is greater, penetrating the upper 30–50 cm of cover soil in general, as reported by Scheutz et al. (2009b), (Figure E.6). The figure also shows the counter gradient of methane gas and oxygen, in conformity with Figure E.5. Other researchers have indicated different ranges of oxidation depths as follows: 15- to 40-cm depth (Visvanathan et al., 1999); 40 to 60 cm (Nozhevnikova et al., 1993); 15 to 60 cm (Barratt, 1995); and 3 to 12 cm (Whalen et al., 1990). These differences are directly related to the conductivities of the soil used in the experiments. Overall, these ranges of depths represent still a shallow range of oxidation in the top cover layer. These ranges must be extended deeper into the oxidation layer and cover layer in order to achieve an effective oxidation barrier.

On the microenvironment level, oxygen concentration can affect the activity of the methanotrophs type I and type II. According to Ren et al. (1997), pure cultures of these methanotrophs are affected by a low oxygen concentration, where the activity is decreased substantially when oxygen level is decreased to below 0.37% (vol/vol), and starts to oxidise methane when oxygen concentration is elevated to the range of 0.45–20% vol/vol. In another research tests involving clay pellets, Gebert et al. (2003) found that no oxidation activities could occur when oxygen concentration is supplied to below 1.7–2.6% (vol/vol) in a passive bio-filter. In contrast, Stein and Hettiaratchi (2001) reported that maximum oxidation would occur at an oxygen level of 0.75–1.6% vol/vol. This contradiction could be explained by the kind of system and type of soil used in both situations.

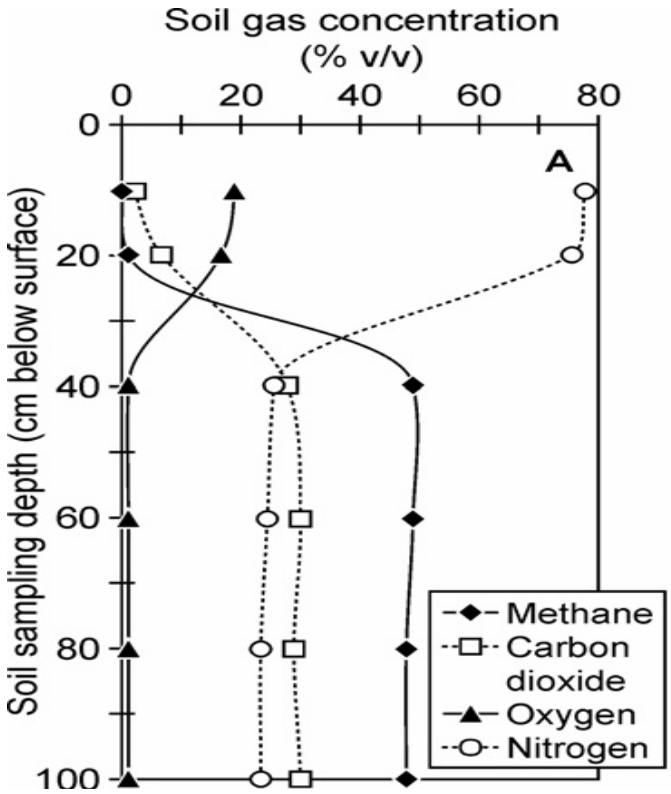


Figure E.6: Distribution of gases through the depth of the soil (source: Scheutz et al., 2009a).

Methane oxidation by methanotrophs occurs in soil layers in the presence of high and low oxygen concentrations, which is due to enzyme catalytic pathways used by these bacteria. Specifically, methanotrophs of type I can perform better in an environment with oxygen concentration of 21% vol/vol. On the other hand, type II has a better performance at a low level of 1% vol/vol. oxygen concentration (Henckel et al., 2000). Although type I methanotrophs perform well for low methane presence, they could only assimilate when methane level is above a threshold of 1% vol/vol (Henckel et al., 2000; Erwin et al., 2005). Furthermore, it has been found that methane oxidation is sensitive to oxygen concentration at lower levels more than that of high concentrations (Czepiel et al., 1996; Stein and Hittiaratchi, 2001). The relationship between methane oxidation and the amount of oxygen concentration present in the soil showed that methanotrophs have limited capacity to use oxygen to oxidise methane. Figure E.7 presents this relationship as investigated by Pawlowska and Stepniewski (2008), using a column test experiment, packed with sand

material. The experiment indicated that methanotrophic activities experienced a slow and gradual increase with the build-up of oxygen concentration from 2.5% vol/vol to 15% vol/vol, then, gradually levelling off in the rate of activities, continuing from that point on to a constant and flat rate, regardless of the increase in oxygen concentrations. Significantly, there is a limited capacity of methanotrophs to oxidise methane in soil, irrespective of the abundance and availability of oxygen to the aerobic bacteria.

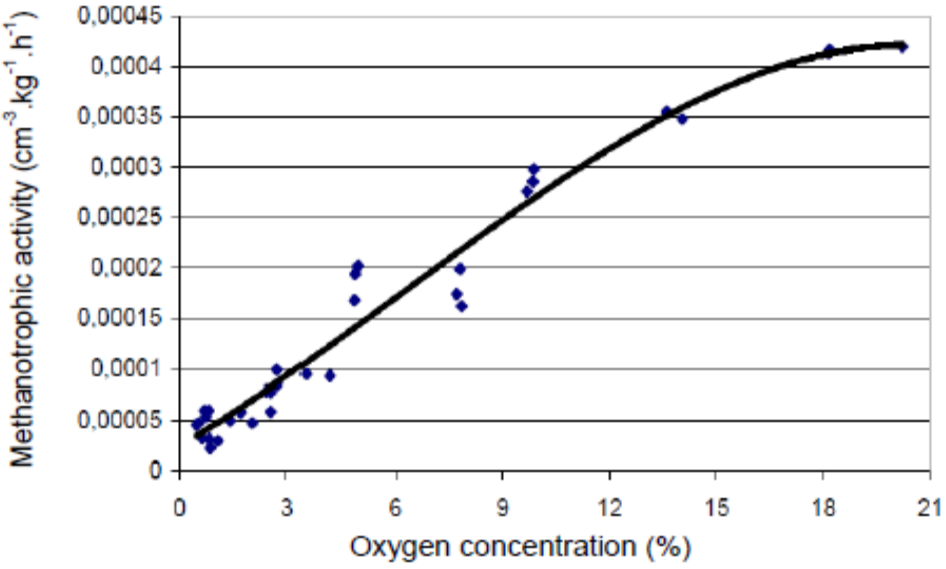


Figure E.7: Effect of oxygen concentration on methanotrophic activities (source: Cao and Staszewska, 2011).

E.11 Methane loading

Methane loading from the lower landfill waste layer upward to the oxidation layers is another important factor affecting the oxidation process. The amount and rate of methane loadings are of particular effects on methanotrophs in the oxidation layer. Landfills generally produce 11 to 17 of landfill gas (LFG) in cubic meter (m^3) of waste per year for newly constructed landfill (10–15 years) in comparison to older landfills, which produce lesser rates at 0.25 LFG per cubic meter per year (Willianson and Bach, 1991). However, these rates are not constant as time elapses on these landfills. These rates tend to decrease from transient to a steady state rates for specific number of years and then tend to decrease further. To better understand the effects of such rates of methane production on the activities of the

methanotrophs, Huber-Humer (2004) constructed a column test with pure methane feed, sufficient oxygen mixing, material of mature compost, maintaining the test at 50% moisture content. The result showed that for 100% methane oxidation, the supply of methane could increase continuously up to a certain range, and then, contrary to expectations, the rate of oxidation could drop sharply, as indicated in Figure E.8. At a methane rate of approximately 200 L/m^2d, the capacity to oxidise methane tends to decrease sharply until it reaches 60% rate of oxidation at approximately 370 L/m^2d. The test however, does not represent landfill gas loading exactly, as the loading in landfills normally consists of 45–60% vol/vol methane to 40–60% vol/vol carbon dioxide. The figure presents the bacterial organisms' activity in the soil of such high methane loadings, indicating that there is a maximum amount of methane loading for maximum oxidation to occur. An investigation conducted by Streece and Stegmann (2003) showed that this maximum amount of loading should be at approximately 60 g of CH_4 per cubic meter per hour ($gHC_4m^{-3}h^{-1}$), for a compost bio-filter, actively fed with landfill gas (LFG) in a mixture of air. Other maximum values of methane oxidation for various soil and environment conditions are discussed later in this research.

In a similar analysis on the rate of activities of methanotrophs, methane concentration was made in abundance and with continuous variation of oxygen as shown in Figure E.7. Powlowska and Stepnoewski (2004, 2006) switched the variable from oxygen variation to methane variation. Results showed an increase in the methanotrophic activities in response to the increase in methane concentration when oxygen was made available in sufficient quantities. Methane concentration continued to rise to a certain constant level; then, these activities levelled off to a constant value from that point on, regardless of the increase in the concentration of methane gas. This was investigated on different levels of soil depths. This behaviour is shown in Figure E.9, and is similar to the nature of the behaviour observed when oxygen was in variation in Figure E.7. All these figures would indicate that the capacity of methanotrophs to oxidise methane, when both oxygen and methane were made available in large quantities, is, to some extent, limited.

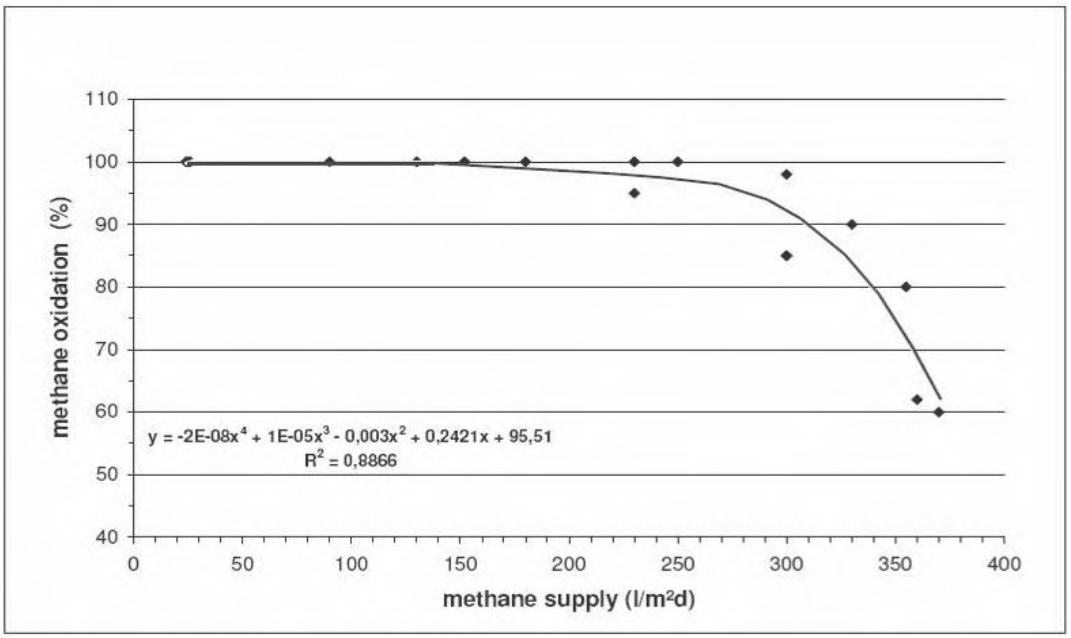


Figure E.8: Methane oxidation efficiency in column tests with pure methane feed (source: Huber-Humer, 2004).

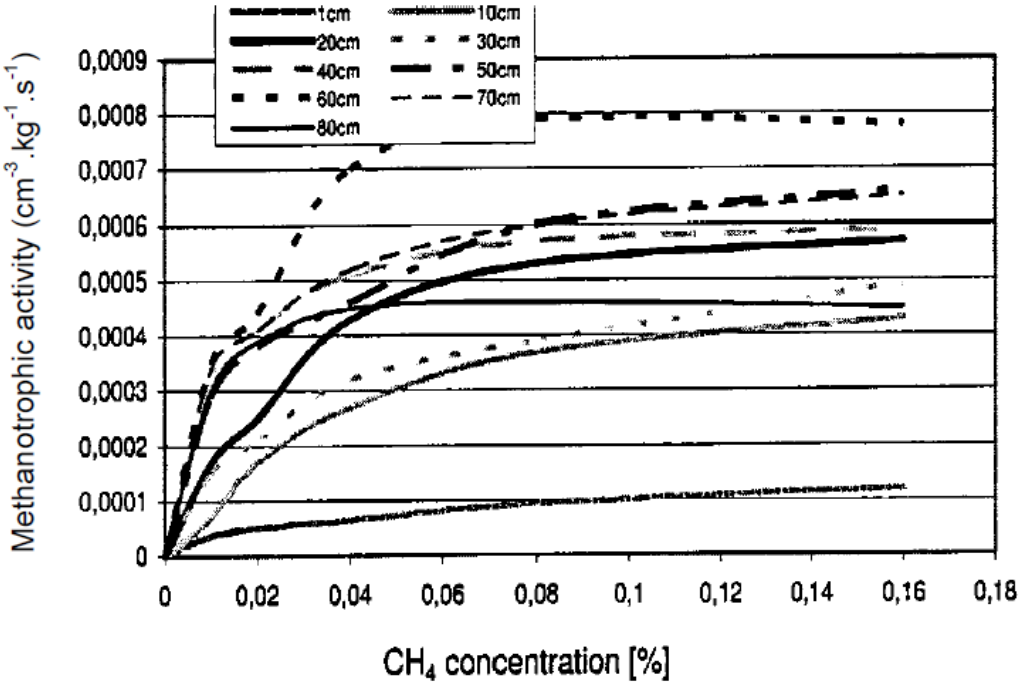


Figure E.9: Effect of methane concentration of methanotrophic activities for different soil depth (source: Cao and Staszewska, 2011).

Molins et al. (2008) investigated the effect of upward methane advective flux on methane oxidation via a column test. They found out that the above air diffusion was decreased by increasing the advective flux due to a rise in the inlet of methane gas inflow. This subsequently changed the pressure gradient, which then reduced the supply of oxygen to a shallower level into the height of the column. This process resulted into a decrease in oxygen supply. Following this, the outcomes of the lower and higher oxygen diffusion were then compared for methane consumptions. As in Figure E.10, the maximum oxygen diffusion at first was up to 0.4 m in depth, and when the pressure gradient across the column was increased from 0.24 m d^{-1} to 4.09 m d^{-1}, the oxygen supply dropped to reach the maximum level of 0.2-m depth. Subsequently, as shown in the figures, the oxidation rate had decreased substantially. Results could indicate that increasing methane advective flux would limit oxidation, which is crucial when designing a bio-cover system.

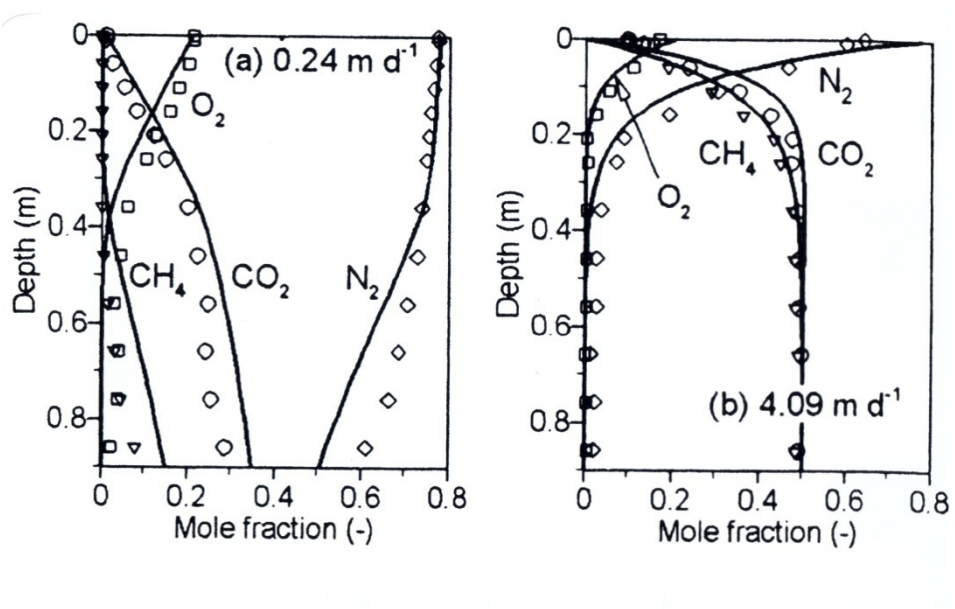


Figure E.10: Effect of advective gases pressure on methane oxidation in a column experiment at CH_4 loadings of (a) 0.24 m d^{-1} and (b) 4.09 m d^{-1} (source: Molins et al., 2008).

Conclusively, from all these studies investigating the relationship between methane and oxygen and the interaction of one over the other, it can be said that the activities of

methanotrophs are dependent on supplying enough oxygen together with methane, for the process of oxidation to continue. If this process were to reach a peak at a certain level of oxygen concentration, alongside with a certain level of methane concentration, subsequently, no efficiency could be had afterwards. Moreover, the activities of the methanotrophs would tend to be confined into a narrow band of horizon in the cover layer at no more than 5–60 cm in depth, regardless of the type of settings or the nature of the environmental conditions arranged in or around them.

E.12 Soil moisture

Water content is another important factor affecting oxidation in landfill soils. Research showed that water content and moisture level in soils have more influence on methane oxidation than temperature (Boecks et al., 1996). In another study, using statistical methods, moisture content was found to be responsible for most of the variations in data collected for methane emission (Christophersen et al., 2000). Soil moisture is important because enough water is needed for the biological activities, nutrient distribution, and waste disposal, and can directly affect the methanotrophic process. Moisture sets the conditions for the growth of bacteria, activation of methane oxidation, and indirectly, affecting gas diffusion (Bender and Conrad, 1995). The diffusion of oxygen into the soil can be hindered; correspondingly, when moisture content is high, the increase of water content can fill in the voids in the soil, thus, limiting the flux of oxygen and methane throughout the soil preventing their interaction. In this latter case, the movement of gas within the soil is substantially reduced, changing the movement from advective and diffusion through air to diffusion through water (Berger et al., 2005). Diffusion through air is much faster and can reach up to an order of magnitude of 10^4 faster than through water (Whalen et al., 1990). Additionally, when water content reaches saturation, methanotrophic activities drop by approximately 56% (Nesbit, 1992). Water content is defined as the mass of water lost from soil when oven dried at 105^{o}C for 24 h divided by the mass of dry soil, and water saturation is the maximum soil holding capacity when soil voids are completely filled with water (US ASTM). Alternatively, the decrease in water content in the soil can also impede the oxidation process by lowering the microbial activities, resulting in methanotrophs' water stress. When water content is

decreased to 13% of the maximum soil water capacity, methanotrophic activities tend to be inactive; zero activities are observed when the water content drops to below 6% of the soil water capacity (Visvanathan et al., 1999). In another study, it was shown that water content could inhibit activities completely when content reached below 32% wt/wt from mechanically biologically-treated soil (Pantini et al., 2015). These situations can happen in desert environments, such as the environment of Kuwait, where rain precipitation does not exceed more than 100 ml a year. Therefore, oxidation is reduced for either the high or low moisture content in soils, leaving the optimum rate somewhere at the middle range, depending on the type of soil and gas flow rates. An optimum level of 13–15% wt/wt of moisture content for upper layers of landfills was reported by Stein and Hettiaratchi (2001) and Park et al. (2002). Others, such as Humer and Lechner (1999) reported 25% to 50% wt/wt for compost material; while Scheutz and Kjeldsen (2004) showed optimum moisture contents to range from 15% to 30% wt/wt for a soil cover.

Other studies have indicated a variety of optimum moisture content for a variety of soil materials and gas loadings (Whalen et al., 1990; Christophernsen et al., 2000; Jackel et al., 2001). Figure E.11 shows the water content profile and its effects on methane oxidation for the upper layers of the soil cover, along with hydrochlorofluorocarbon gases (Scheutz and Kjeldsen, 2004), indicating that water content in soil has a profound impact on methane oxidation, and shows a zero oxidation rate when moisture content reaches below 6% wt/wt or above 50% wt/wt. The difference in these estimates could be attributed to the microstructure of the medium that has been tested. This is indicative of the amount of moisture content inside the pore spaces, along with the sizes of grains, which have provided a greater surface area for the bacteria to multiply, thus, affording a likely chance of affecting the transport of nutrients to the bacteria. In addition, each soil medium, because of its own distinct physical and chemical makeup, could contain salts, biological matter, other competing species of bacteria, and chemical components, consequently, altering the chemical nature of the moisture content present in the soil, eventually, to affect the oxidation profiles.

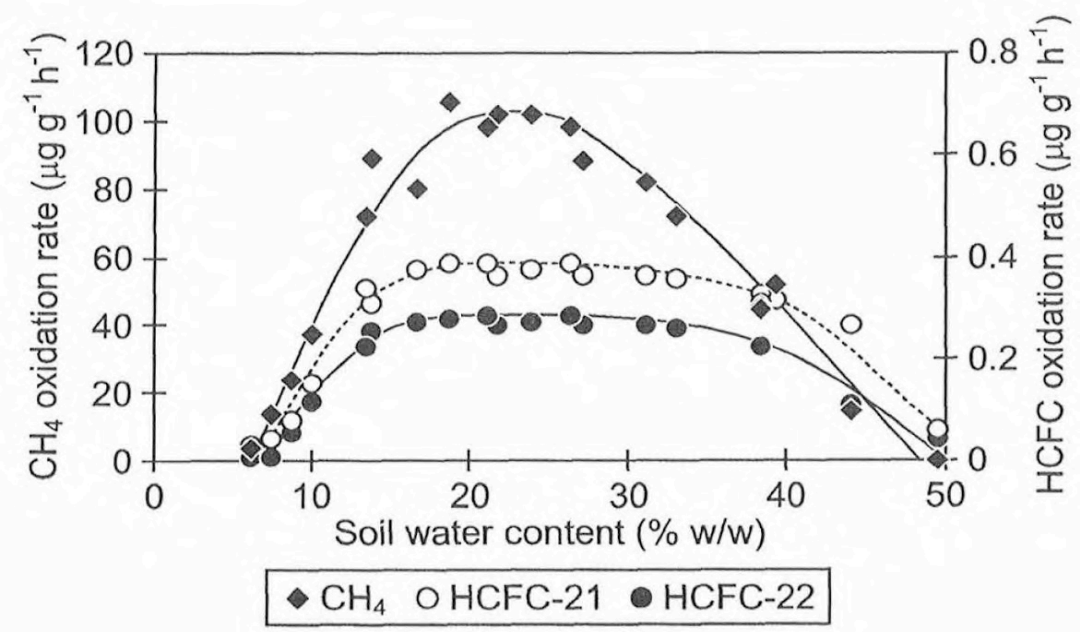


Figure E.11: Moisture content and its effect on methane and hydrochlorofluorocarbon oxidations in landfill soil cover (source: Scheutz and Kjeldesen, 2004).

Moisture available in soils depends on soil grains sizes, structure, pores sizes and pores alignments, water surface tension, capillary effect, and the interacting minerals present in the soil. Several standards are available to measure the water content quantitatively in the soil. The use of these standards is dependent on field of application, where gravimetric dry moisture content (MC) (also called absolute or natural water content, or just moisture content), measured as the ratio of weight of water to the dry weight of the soil, is the standard mostly used in the field of landfill analysis (Figure E.11, Table 2.5, Table 2.6). This standard is used in this study for comparative analysis with literature data. Other standards, such as field capacity (FC), permanent witting point (PWP), available water capacity (AWC), residual water content (RWC), total water capacity (TWC), and several other specialized standards are used for other fields of application, where such applications are soil analysis, hydrology, agriculture, and irrigations. Relationships between the parameters of these standards are studied extensively and available in published literature (Karube and Kawaï, 2001; Miller et al., 2002; Sugii et al., 2002; Stein and Hettiaratchi,2001; Park et al., 2002; Scheutz and Kjeldsen, 2004).

E.13 Nutrients

Nutrient elements in soil are important to the methanotrophic activities. These elements, when added through water in experimental procedures, or when present naturally in soils, are essential in enhancing cellular metabolism. While some of these elements could encourage methanotrophic action at specific range of concentrations; albeit, at other amounts, they could likewise inhibit activities, and could even prove to be toxic for the bacteria. Nutrients such as nitrogen and compounds, copper, phosphate and phosphate compounds, and other nutrients, discussed in the following sections, are elements known to somehow affect methanotrophic activities.

E.14 Nitrogen and compounds

Inorganic nitrate provided in the soils in the form of nitrate ions (NO_3) in general, has no significant effect on oxidation. However, it can inhibit methanotrophic activities at high concentrations (Bodelier and Laanbroek, 2004). The source of these nitrate ions often comes from sodium nitrate, potassium nitrate, or from ammonium nitrate, when added in soils through organic or inorganic fertilizers. Mostly, the nitrate is used as a source of nitrogen fixation (Le Mer and Roger, 2001).

The most studied nitrogen element in the literature however, is ammonium (NH_4^+), as it is added in the soil in the form of ammonium chloride, ammonium sulfate, or from urea, as amendments. This nutrient element could simulate oxidation if the prevailing conditions, such as the pH values, methane concentration, and the type of bacteria present, are right. Studies in the literature showed that high concentrations of ammonium could encourage converting NH_4^+ to NO_2, resulting to oxidation being inhibited, as ammonium competes with methane for oxygen$^-$ (Novikov and Stepanov, 2002; Boecks and Van Cleemput, 1996). The studies showed that 12–28% of the bacterial population is engaged in this conversion. The amount of inhibition was shown by Boecks and Van Cleemput (1969) to range from 25 mg-N/kg of cover soil, and would decrease linearly with the increase of NH_4^+. Conversely, Scheutz and Kjeldsen (2004) showed that the oxidation of methane is not affected, up until

reaching 14 mg-N/kg (and HCFC compounds), then rapidly decreasing with a continuous increase of NH_4^+ as shown in Figure E.12. Other studies however gave different results. Hutsh (1998) indicated that an amount of 40 mg-N/kg showed marked inhibition when ammonium chloride is added to arable soil, inhibiting oxidation completely. Other studies have shown that bacterial activities could stop, when the ammonia were to increase to 1000 mg/L, although however, survival of the microorganisms was not affected (Liu et al., 2015). The difference in these results could be attributed to the amount of oxygen available in the test bed, where the inhibition is dependent on the degree of competition between bacterial species for the available oxygen.

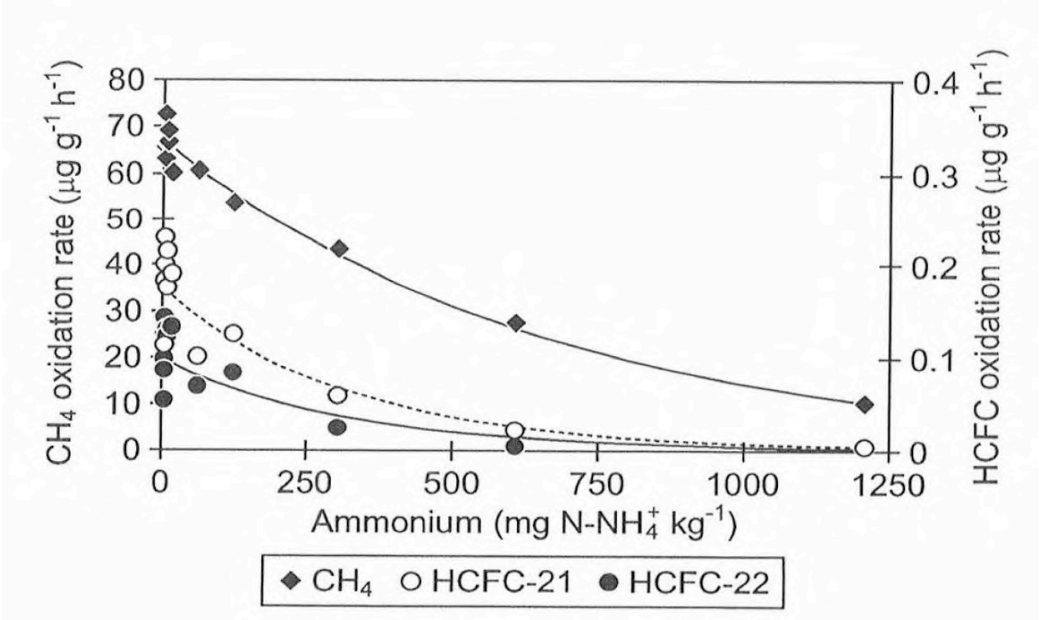


Figure E.12: Methane oxidation rates due to variation in ammonium and HCFCs (source: Scheutz and Kjeldsen, 2004).

Methanotrophic bacteria can assimilate nitrogen from the atmosphere directly, especially methanotrophs type II, which is a process that can take place when nitrogen is depleted from the soil (Hanson and Hanson, 1996); although, this process takes up more energy. This allows the other processes of assimilating nitrogen from the inorganic materials (nitrogen fixation), existing naturally, more reliable than those which are added to the soil, or taken up from the atmosphere. The other type I methanotrophic bacteria do not have the ability to fixate nitrogen; although, they could respond to the deprivation of nitrogen by producing extra cellular polymeric substance (EPS) (Wilshusen et al., 2004b). Methanotrophic bacteria

have been found to have high requirement for nitrogen, as they need one mole of nitrogen for every four moles of carbon (Anthony, 1982), up to certain concentrations. When the molar ratio of carbon to nitrogen (C/N) exceeds 10; subsequently, nitrogen becomes limiting (Bodelier and Laanbroek, 2004), which is often the case in landfill soil, as methane is more abundant than nitrogen. Additional limitation on methane oxidation might occur when vegetation is irrigated on top of landfill soils, due to the high uptake of nitrogen from the soil by plant roots (Bodelier and Laanbroek, 2004). The effect of nitrogen on methane oxidation and on the activity of methanotrophic bacteria, however, can be quite complex. Often, published information is contradictory. Hence, no clear trends or consistent conclusions can be drawn from published studies. Table E.5 shows some results obtained from the literature with variable outcomes (Nikiema et al., 2007).

N forms and Concentration	Effect	Filler Beds	Sources
25- mg N/kg soil in the form of NH_4^+ or NO_3^-	Improved CH_4 elimination by 100%	Soil	Hettiaratchi et al., 2000
≥ 30 -mg N/kg soil in the form of NH_4^+ or NO_3^-	Inhibiting CH_4 elimination	Soil	Chiemchaisri et al., 2001
10- to 200- mg N–NH_4^+ /kg soil	Inhibiting CH_4 elimination, however, its extension depends on the type of soil	Soils	Bronson and Mosier, 1994; Cai and Mosier, 2000; Hettiaratchi et al., 2000; Novikov and Stepanov, 2002; Park et al., 2002
Sodium nitrate, from 0.14- to 0.75- g N/L	5 times increase in the elimination capacity (from 130 to 700 g/(m^2. d).	Inorganic filter material	Nikiema et al., 2005
Sodium nitrate > 0.75- g N/L	Decrease in the CH_4 oxidation	Inorganic filter material	Nikiema et al.,2005
25–to 200- mg N– NO_3^- /kg soil	No CH_4 elimination effect	Soil	Beckx and Van Cleemput, 1996; Park et al., 2002
2,500- mg N– NO_3^- /kg soil	Inhibiting CH_4 elimination	Soil	Kumaraswamy et al., 2001

Table E.5: Studies indicating the effect of nitrogen on methane oxidation (Source: Huang et al., 2011)

In arid land environments, plants and small vegetation are rare; therefore, organic nitrogen is limited, leaving the bacterial community dependent on inorganic compounds present in the soil. Depletion of the small amount of organic matter that could be present during landfill covering from organic matters and the inorganic substances have an adverse effect on methane oxidation. Imperative is to add compost or similar material rich in nitrogen with ammonia or sodium nitrate compounds in order to offset this lack of nitrogen nutrients.

E.15 Copper

Copper is an important element for microorganisms, plants, and animals alike, to build up different proteins. It is present naturally at concentrations of 0.5 ng/m^3 in the atmosphere and ranging from 2.0 to 40 mg/kg in sediments, but it could reach far higher ranges in contaminated soils. Among copper manufacturing plants and in soils treated with fungicides based on copper, copper could reach up to 1000 mg/kg soil (Scheffer et al., 2002). Copper (Cu) in soil is present in forms of chalkosin (Cu_2S), chalcopyrite ($CuFeS_2$), and copper oxides such as malachite ($Cu_2(OH)_2CO_3$), or as a free copper (Scheffer et al., 2002).

Copper is a nutrient additive that affects the growth of bacteria, enhances their growth activities at specific concentration, and inhibits them at different levels of concentrations. Hanson and Hanson (1996) indicated that copper could obstruct the action of sMMO enzyme at concentration of 1 µmol/L; while it stimulates synthesis of pMMO enzyme at concentration of 1–5 µmol/L, meaning in that, bacteria metabolise methane using either process, depending on the concentration of Cu. For higher concentration of Cu, cells use pMMO enzyme, as well as the sMMO enzyme when copper is limited in its environment. Other studies showed that methane oxidation could be increased slightly to about 5%, when adding a copper compound, $CuCl_2$, to a paddy soil at a rate of 0.02 g/kg of soil (Mohanty et al., 2000). On the other hand, , Bender and Conrad (1995) found that oxidising methane had ceased completely, when copper concentration was increased to 4.3 mM, suggesting that this range is the limit for complete inhibition. This would infer that the presence of copper is

essential and is needed for the active functioning of the methanotrophs. Nevertheless, its concentration must be maintained to a moderate level.

E.16 Phosphate and phosphate compounds

Phosphate is another nutrient needed for the growth of methanotrophs. Yet still, research published in the literature showed little evidence of any enhancement of methane oxidation, when methanotrophs are provided with phosphate compounds. Le Mer and Roger (2001) showed that when adding 0.1 g of P-K_2HPO_4 /kg of soil as a nutrient, the effect on methane oxidations was found to be negligible. Sinke et al. (1992) confirmed through a batch experiment that the growth of methanotrophic bacteria contributes to the phosphate (P) uptake of aerobic sediment, calculating that a molar ratio of 45 carbons to phosphate has contributed to the growth of methanotrophs with little evidence of changing the rate of oxidation. A more recent investigation carried out by Zheng et al. (2013) discovered that addition of P to a paddy soil has reduced methane oxidation; but otherwise, enhancing the abundance of methanotrophs, suggesting that adding phosphate fertilization to the soil should be done with caution. Thus, the role of phosphate and phosphate compounds on methane oxidation as yet, remains to be unclear and has to be added carefully in the soil.

E.17 Other nutrients

The effect of iron as a nutrient was studied by Boiesen et al. (1993), and was found to have similar effects on methanotrophs as that of the phosphates, increasing the protein formation. The study showed that a yield of 0.49 mg of protein is produced per one milligram of methane, when iron was added at a concentration of 0.10–5.0 mg/L. The study also showed that iron addition has increased methane utilisation rates. Effects of potassium sulfate and manganese oxide on methane oxidation had also been investigated by Kumaraswamy et al. (2001). Findings showed a marked increase; while high concentration levels of sodium chloride and potassium chloride were seen to inhibit oxidation (Kravchenko, 2002; Gebert et al., 2003).

Nutrient elements in soil are important to methanotrophs and essential to their existence and growth; however, the results obtained from the literature were varied. Most elements enhanced oxidation up to certain concentration levels; while others proved to inhibit oxidation, and even showed that they could be toxic at higher levels of concentrations. The reason for these diverse findings could be attributed to the nature of soils used, type of experiment carried out, type and amount of nutrient applied, etc. In other words, there are no common standards for comparison of these studies. Furthermore, studies which have been carried out concerning nutrient elements each, singly added to the soil in an experiment did not in all reality represent the actual and natural makeup of the soil in landfill. In nature, soils have numerous nutrient elements that interact with each other and with the methanotrophs and with other multitudes of coexisting bacteria, which could thus result in a nonuniform or inhomogeneous outcome.

E.18 Temperature

Like all other biological beings, cells react directly to variations in the temperature; among them is the reaction of methanotrophic bacterial cells. This reaction is profound when the bacteria assimilate methane in an environment controlled by the temperature, greatly affecting the oxidation process. Borjesson and Svensson (1997) reported that soil temperatures could be responsible for 85% of the measured oxidation variation of methane. Methanotrophic bacteria flourish in certain range of temperatures, where they continuously function up to a temperature of 36°C; and then, the oxidation process stops completely at 45°C (Czepiel et al., 1996). These high temperatures could pose a dilemma for the desert environment. On the other side of the scale, oxidation rate is very limited at a temperature of around 4°C (Humer and Lechner, 2001); although, significant methane oxidation could be observed at lower temperatures, i.e., 2–15°C. Christophersen et al. (2000) found methanotrophic activities ranging from 0.04 to 0.017 (μmol/g h) at 2°C and 0.34 to 1.17 (μmol/g h) at 15°C. Other researchers have found similar results (Kettunen et al., 2006; Cabral et al., 2007).

For optimum values, the temperature varies from one range to another, as indicated from published literature. Boecks et al. (1996) showed an optimum range of 20–30°C. Borjesson and Svensson (1997) put the optimum range at 25 to 35°C. Nesbit (1992) showed an optimum of 20–30°C; while Whalen et al. (1990) reported an optimum value at 30°C on landfill cover soil, and Humer and Lechner (2001) indicated an oxidation rate of 70–80% at 18°C. Figure E.13 shows typical effects of temperature variations on methane oxidation for batch experimentation (Whalen et al., 1990). An interesting finding was reported from an investigation carried out by Czepiel et al. (1996), where they found increasing oxidation rates when the temperature was increased; although there was no apparent optimum oxidation range, as indicated in Figure E.14. Consequently, from all of these studies, the optimum oxidation rate due to temperature could be positioned at the range of 18 to 36°C, depending on the type of soil being used, notwithstanding, the findings of Czepiel et al. (1996).

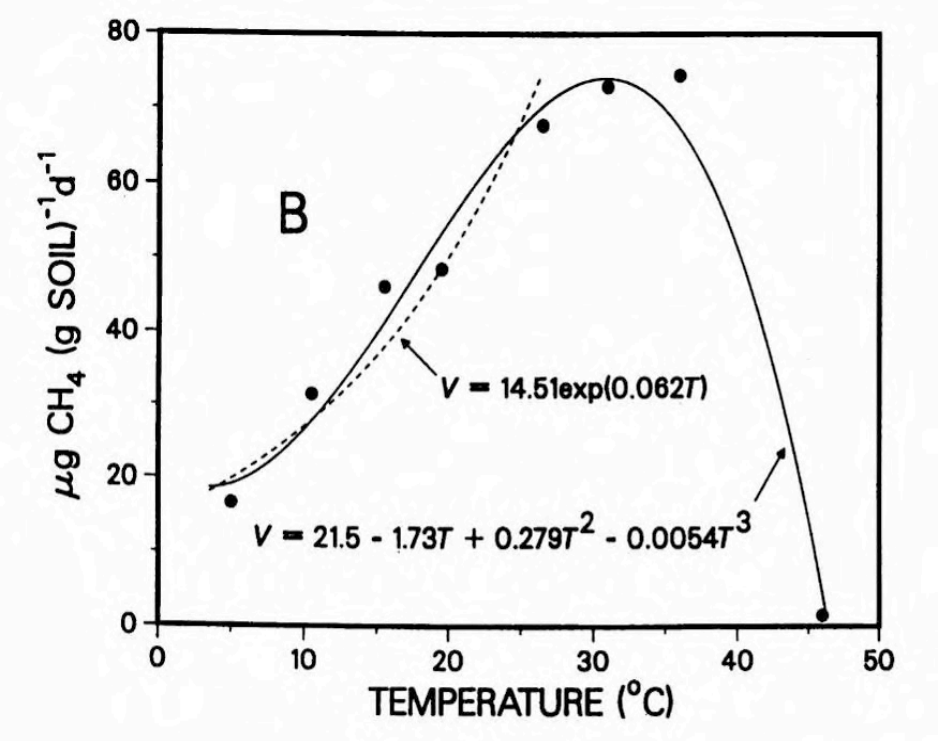


Figure E.13: Effect of temperature on methane oxidation (source: Whalen et al., 1990).

For the type of methanotrophic bacteria, it was observed that methanotrophs type I dominate at low temperatures of less than 10°C; while type II methanotrophs were seen to be dominant at temperatures of 20°C and higher. Accordingly, a mix of both types could be

active in the soil, if the temperatures were to be in between these ranges (Borjesson et al., 2004). This observation would indicate that temperature is a selecting factor, dictating which species to act in the conversion of methane in landfill soils.

While temperature is the most controlling and dominant factor among all others, moisture content in soil could limit oxidation, as previously explained. To establish this fact, Boecks et al. (1996) discovered in an experiment that variable moisture content could affect oxidation in a landfill cover soil. But then again, the effects were fairly weak and varied only slightly.

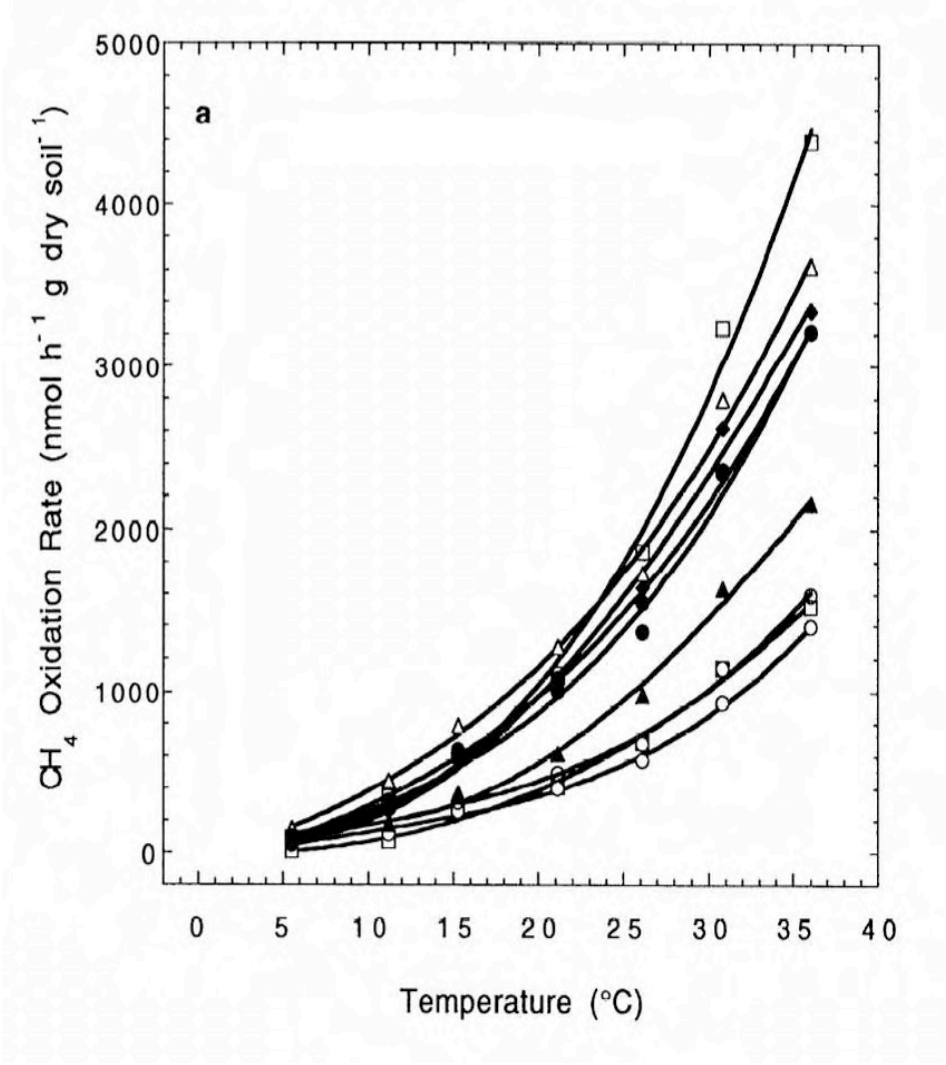


Figure E.14: Influence of temperature on methane oxidation of several landfill cover samples (source: Czepiel et al., 1996).

E.19 Acidity of the soil

The pH characteristic of the soil media is of relevance to the activities of the methanotrophs and of the microbial oxidation process. The investigations revealed that when the acidity of the soil medium is increased up to the range of < 4.5, or the pH is increased to > 8.5, the oxidation process is substantially decreased. Bender and Conrad (1995), Huber-Humer (2004), and Scheutz and Kjeldsen (2004) indicated that methanotrophic bacteria perform best in soil media when the pH range is between 5.5 and 8.5, as presented in Figure E.15 (Scheutz and Kjeldsen, 2004). The curves in the figure show that methane oxidation, as well as hydroflourocarbons, do change with the variation of the acidity of the media, all in a similar fashion, suggesting that methanotrophs prefer a neutral soil background of pH 6–7. These values are obtained through lab experimentations. However, in an *in situ* investigation, Bender and Conrad (1995) found out, that when investigating four soil types, forest soil prefers a medium of pH 4.5; while cultivated soil has an optimum of pH 8.1. Nevertheless, the average optimum pH values of all the four soils investigated were known to be in line with the experimentation values, averaged at 6.7 to 7.5. In general, the pH values for the growth of most methanotrophs can be put at 6.6 to 6.8, as suggested by Whittenbury et al. (1970).

While the acidity of soils is of relevance to methane oxidation in natural settings, controlling the acidity is neither guaranteed nor maintained. Changes in pH values are dynamic in soils, and any chemical additives to alter their acidity are washed out with the diffusive water and with the seepage of leachates. In addition, soils in general, have a high buffer capacity that can lessen the changes in the acidity (Scheutz and Kjeldsen, 2004). Consequently, the effect of pH on the growth and the activity of methanotrophic bacteria was determined to be not highly significant.

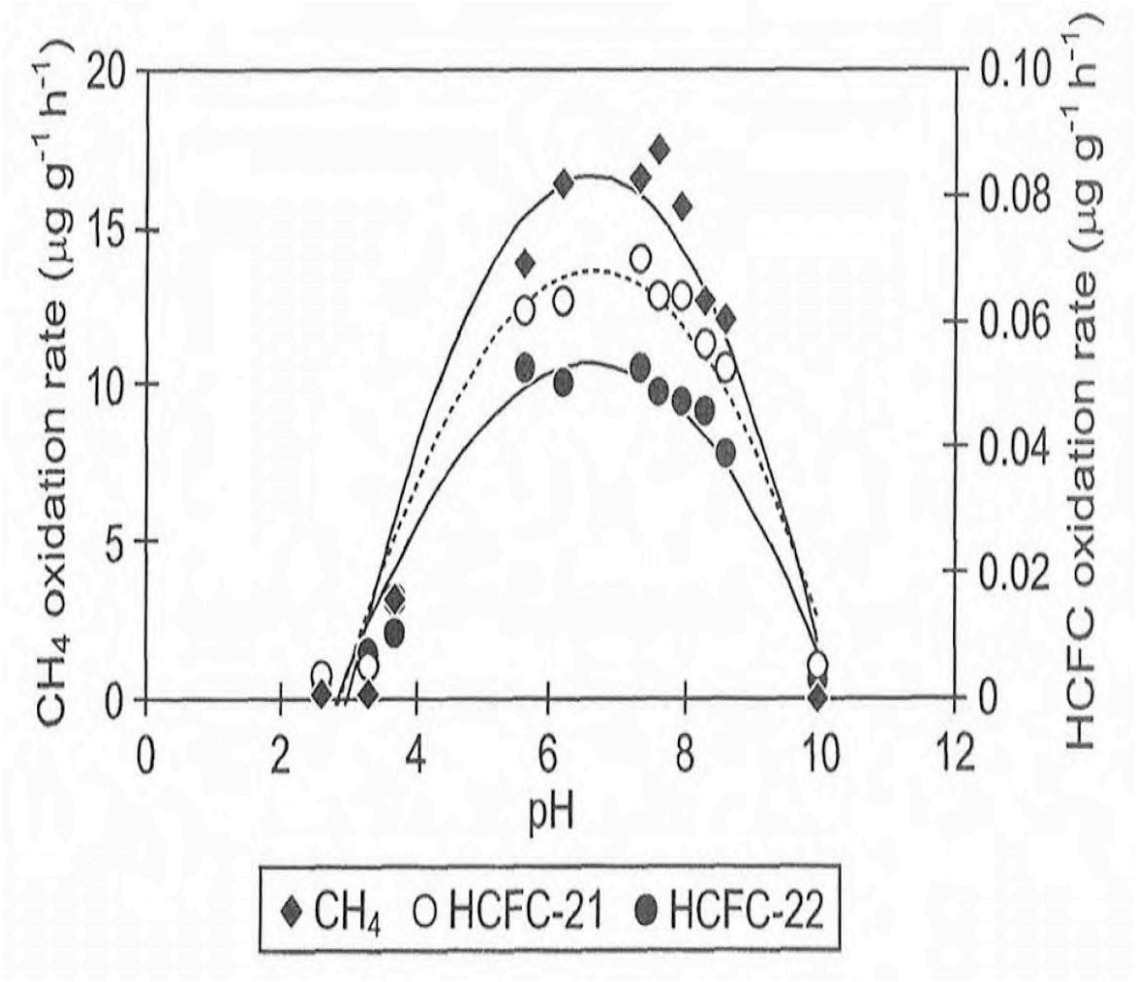


Figure E.15: The effect of pH acidity on rate of microbial oxidation (source: Scheutz and Kjeldsen, 2004).

E.20 Atmospheric pressure

Oxygen is an essential element for the synthesis of methane in landfills, as explained in Section E.10; however, its availability and continuity are of greater importance. In the natural setting of landfills, oxygen is available in voids and cracks of the layers, trapped, when landfill sites were first established, and equipped by means of diffusion or advection. The diffusion of oxygen into the activity cover layers is driven by the changes in concentration of gases and that in turn, depends on the characteristics of the soil, as discussed in Section E.10. While the advective flux of oxygen is propelled by pressure gradients, the pressure changes with the elapse of time over the depth of the cover layers and with the changes in

barometric pressure above the layers. This latter advective transport is induced by the wind fluctuation and the atmospheric pressure (Kjeldsen and Fischer, 1995).

Barometric pressure, as a mechanism of delivering oxygen, was investigated by several researchers (Kjeldsen and Fisher, 1995; Christophersen and Kjeldsen, 2001; Nwachkwu and Anonye, 2013; Xu et al, 2014), who determined that there exists an inverse relationship between pressure and gases emitted from landfills. When pressure is higher above the ground level, methane, carbon dioxide, and all other volatile gases emitted from the landfill tend to lower their presence in the atmosphere and *vice versa.* Figure E.16 shows a typical atmospheric pressure/methane soil gases behaviour for a case of methane emitted from Bluff Road landfill near Lincoln, Nebraska, USA, in 2010 (Xu et al., 2014), in which emission had subsided when confronted with cold front high pressure level (dashed line). It is also worth noting from the graph that methane emission from the landfill had been suppressed close to zero, but not relatively reaching that level, and that, there was no inverse methane injection to the soil.

The response of cover layers to the rise or fall of barometric pressure depends on the prevailing pressure inside these layers. When gas pressure inside the layers is higher, oxygen delivery to the inside of the layers decreases substantially, as the advective flux of gases is much higher, hence, overpowering the pressure gradient above the ground (Christophersen et al., 2001). However, for old small landfills and landfills with low gas emission, atmospheric pressure could have some effect on transporting oxygen to within the reaction layers of the landfill. On the whole, pressure inside the layers and pressure above ground would tend to balance each other, as a continuous monitoring of unlined and unmonitored landfill for the recovery of gases has been indicated. This monitoring showed that a harmonic and in phase with the above ground barometric pressure balance each other, and that, there would only be a small difference when the landfill is dry or wet (Bonger et al. 1987).

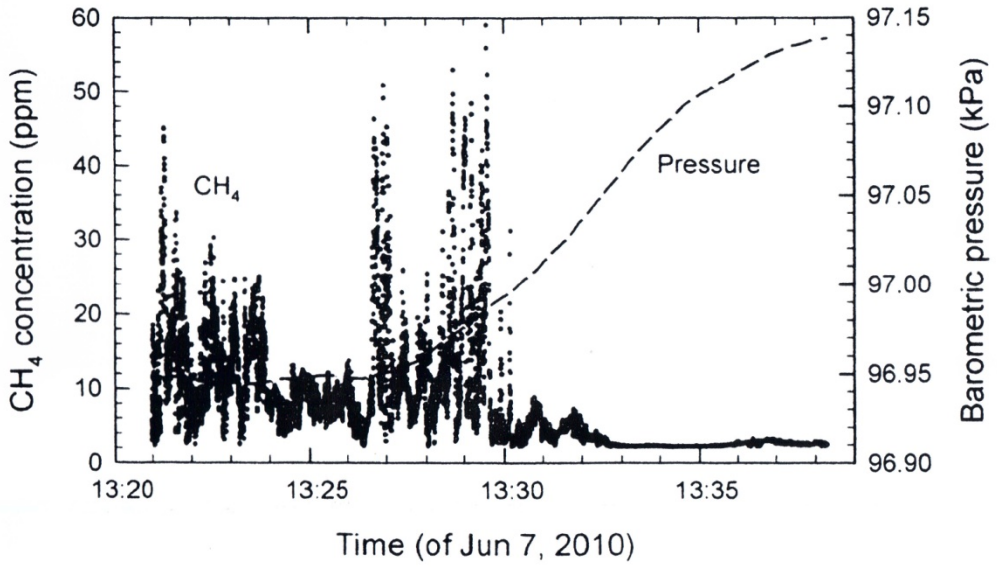


Figure E.16: Effect of pressure on methane emission from Bluff Road landfill near Lincoln, Nebraska, USA in June 7, 2010 (source: Xu et al., 2014).

The pumping mechanism for oxygen delivery into the landfill soils is indicated by the effect of barometric pressure on landfills. However, recent monitoring of existing landfills showed that barometric pressure fluctuations ranged around +/- 0.1 KPa h^{-1} for a half-year time span average (Xu et al., 2014). These are small variations of pressure to have caused noticeable air infiltration into the soil. Moreover, soil conditions when wetted with water, filling the gaps and pores of the soil, could even impede the effectiveness of atmospheric pressure in delivering oxygen to the oxidising bacteria inside the reaction layers. Another *in situ* study conducted by Nwachykwu and Anonye (2013) on a landfill in Manchester, UK, to monitor atmospheric pressure on methane and carbon dioxide, indicated that above ground pressure is insignificant in ground-gas control. In agreement, Christophersen et al. (2001) observed that the barometric pressure does not appear to be the most controlling factor in landfill gas emission, which was similarly observed by Borjesson and Svensson (1997) in their investigation of a Swedish landfill. This outcome could be attributed to the fact that the atmospheric pressure has the characteristic to change continuously on a short time frame;

and conversely, gas pressures inside the soil could change slowly over a longer space of time. Soils are different mediums from the atmospheric medium, and the soil medium has a makeup consisting of different materials and filled with varying degrees of moisture. Although some correlation can exist between barometric pressure and the free methane emission to the atmosphere and the environment, as already discussed; albeit, this relationship does not control the air or methane diffusion into the soil. Xu et al. (2014) observed from the data collected from the Bluff Road landfill along with the data presented by Shurpali et al. (1993) and Tokida et al. (2007), that the emission from or into the landfill does not depend on the absolute value of the barometric pressure; but rather, on the gradient of the pressure (dP/dt) inside the soil layers and on the prevailing atmospheric pressure. When the pressure inside the landfill is higher, as the case among fresh landfills, methane and other gases will be emitted into the atmosphere, regardless of the changes in the atmospheric pressure, inferring that this is due to the advective higher pressure inside the soils and would prevent fresh air from filtering in (Xu et al., 2014). Therefore, atmospheric pressure fluctuations may be said to have a weak or insubstantial influence on providing fresh air (oxygen) into the reaction layer of the methanotrophs.

E.21 Wind turbulence and speed

Although wind turbulence can induce pressure gradient immediately on the top soil surface of landfills; albeit, the soil's physical characteristics can affect the level and degree of pressure gradient in the soil, due to these fluctuations. Besides the physical properties of soil, mentioned in Sections E.3-E.9, surface characteristics, such as plants, irregularities on soil surfaces, and surface grain shapes and sizes are also of relevance to the wind-induced static pressure (Waddington et al., 1995). When wind is blown across the top layer surface, irregularities in the physical surface, as well as the texture of the grains, force air to move up or down in that microenvironment, leaving the air particles to move quickly. This quick movement creates micro vacuum pockets across the soil surface, allowing an exchange of air between the atmosphere and the soil beneath it. This wind-induced mechanism is a diffusive transport action strongly influenced by permeability, porosity, and water content of the soil.

Constant wind speed blowing over for extended periods could affect the barometric pressure, as the case for major weather occurrences. Short and fluctuating wind speeds blowing over the terrain are the normal weather daily conditions. Such weather conditions have the potential to create a continuous driving pressure fluctuation along the tangent surface of the landfill with the atmospheric medium, thereby, affecting the advection of oxygen into the reaction layers of the landfill (Ishihara et al., 1992). Poulsen and Moldrup (2006) proved in their research of Skellingsted landfill in Denmark that the magnitude and penetration of pressure attenuation inside the top surface layer depend basically on the standard deviation and the power spectrum of the pressure fluctuation just above the soil surface. In turn, the standard deviation and the power spectrum are dependent on wind turbulence, and which is dependent upon the wind speed and the surface roughness of the soil. They also found that wind-induced pressure is the most important mechanism in transporting air into the soil, but which relies largely on the physical characteristics of the soil, such as porosity, permeability, and water contents. They also determined that gas emission from landfills due to wind turbulence could count for up to 40% of all other pressure forces responsible for gas transport. Conclusively, the same authors said that wind-induced pressure transporting gas into or out of the soil could be the most effective factor when soil is saturated with water, as it could better reduce diffusivity compared to air permeability (Poulsen and Modrup, 2006). These findings could be accounted for when using mathematical simulation models, as wind-induced pressure, variation sequences, with measured standard deviations, and power spectra are difficult to measure naturally due to the nature of wind randomness and turbulence.

Investigations in the literature of diffusive and advective air transport, as a mechanism of exchanging gases and vapour between the soil and atmospheric media are well-established, as discussed in the section of soil properties. However, investigations into the effects of wind speed fluctuations on oxygen transport into the soil are scarce and inadequate. Besides the research done by Poulsen and Modrup (2006) of wind speed fluctuation and interaction with soil, Huber-Humer et al. (2008) investigated the effect of wind speed on compost bio-covers placed on slopes, and concluded that wind has a great influence on these particular covers, predominantly, since the covers possess the characteristics of a high degree of permeability.

Also, Takle et al. (2003) investigated the frequency of induced atmospheric pressure variation of up to 2-Hz fluctuation on the flux of gases in a field soil. They reported that at the frequency of 2 Hz, the fluctuating pressure could easily penetrate the soil to a depth of several centimeters. It is worthy to mention that the experiment was performed on a relatively dry field. Therefore, wind energy and its fluctuating speed over an irregular terrain can produce significant amount of pumping pressure over landfill soil with enough energy to penetrate soils to a depth of 0.60 m for dry soil and as little depth as 0.065 m for wet soil, for a wind fluctuation frequency at the range of 0.25 Hz (Takle et al., 2003). Figure E.17 shows the pressure variance with passing time for 15-, 45-, and 60-cm depth beneath the soil surface level, indicating that the pressure behaviour for these depths is similar and is in phase with each level of depth, reaching up to a pressure amplitude of 3 Pa when measured at a wind frequency of 2 Hz.

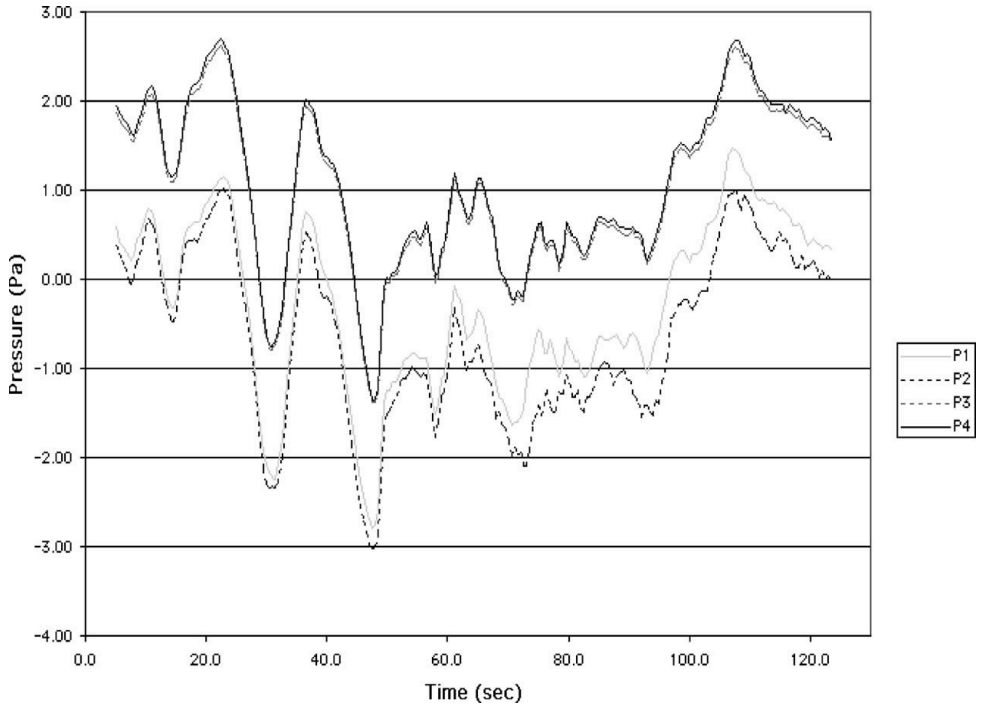


Figure E.17: Pressure fluctuation vs. time at depths, P1= 15 cm, P2= at the surface, P3= 45 cm, P4= 60 cm, at 2- Hz wind speed frequency (source: Takle et al., 2003).

E.22 Oxidation inhibiting substances and elements

Methane oxidation inhibition can be influenced either by the soil characteristics and composition, or by the type of substances and elements present or added to the soil. Inhibition could also be influenced either by interfering with the assimilation process by binding with the enzyme of the cells, or by a complete toxicity of the enzymes themselves (Scheutz et al., 2009a). Soil characteristics include the soil's micro arrangements of grains and voids that affect the population of the methanotrophs. Their composition and location within the soil also affect the inhibition pattern of the activities of the methanotrophs, as observed by Visscher and Van Cleemput (2003); while toxic additives or toxic components present in the soil have been observed to damage the cells of the bacteria irreversibly.

E.23 Inhibiting chemical elements

Studies on landfill cover soils showed that chemical elements could inhibit methane oxidation completely, due to their strong toxic effects on the cell of the bacteria. These chemicals and their effects are summarized in Table E.6; although, others may also exist, such as pesticides and fertilizers used on soils. Generally, oxidation depends greatly on the concentrations of the substances that were there in the soil in the first place and on the elements-added to the soil. These substances influence the characteristics of the soil surrounding the microenvironment of the methanotrophs and influence the concentration of methane present in the same environment.

Inhibitor	Inhibition	References
C_2H_2 C_2H_4	0.01 ml/L for inhibition 1 ml /L for inhibition	Chan and Parkin, 2000
HNO_3	Strong inhibitor	Bradford et al., 2001
NH_4^+/NO_3	>µ30g/g dry soil	Chiemchaisri et al., 2001
Acetylene/ethylene	Strong inhibitor	Prior and Dalton, 1985
Copper	>60 mg/Kg of soil > 4.3 mM	Scheutz and Kjeldsen, 2004; Bender and Conrad, 1995
Difluoromethane	Storng inhibitor	Miller et al., 1998
Dichloromethae	Strong inhibitor	Byers and Sly, 1993
Methyl fluroride	Strong inhibitor	Frenzel and Bosse, 1996
Pesticides: Oxadixyl, atrazine, dimethenamid	Inhibitors	Boeckx et al., 1998
Methanethiol/carbon disulfide	Inhibitors	Borjesson , 2001

Table E.6: Some methane oxidation inhibiting elements

E.24 Physical inhibitors

The flow of gases through the oxidation layer and the cross encounter of each other along the depth of the layer is a complex process. For advective or diffusive fluxes of gases into the oxidation layer, the rite of passage could be an inhibiting process by itself. When a high rate of methane gas moves upward from the waste zone, it forms a high gradient zone, consequently, pushing and preventing oxygen from diffusing downward into the layer (De Visscher et al., 1999), and therefore limiting the oxidation. In addition, the physical stability of the soil itself could have a constraining factor on oxidation. A compost cover layer when fresh would have more oxidation activity than when it becomes old and has endured settlement. This action of settlement would restrict availability of oxygen, hence, limiting oxidation. Scheutz et al. (2011) found that an eight-year-old settled compost of garden waste had a methane oxidation rate of 4.8- µg h^{-1} CH_4/g of dry mass (DM) compared to a

four-year fresher garden waste that oxidised 161-μg CH_4- g DM h^{-1}. Additionally, the degree of soil compaction can have a significant effect on grain alignment and void space volume, therefore, affecting porosity and the ability of air to diffuse into the soil, and hence, inhibiting oxidation. Natural soil compaction could amount to 20% for compost cover down from its original volume, as Huber-Humer et al. (2008) had found. Moreover, the physical makeup of soils, when they are rich in nutrients could encourage the presence of diversified microorganisms; consequently, each microorganism species will compete for the available air, as air is the only source of oxygen. This action therefore, has an inhibiting effect on methane oxidation (Kettunen et al., 2006).

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